Ultrafast laser micro and nano processing with nondiffracting and curved beams

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Abstract

Ultrafast laser materials processing has undergone an important change with the development of non-diffracting beams. These beams enable overcoming many of the difficulties usually encountered with standard Gaussian-beam focusing in materials. We review the techniques of non-diffracting and accelerating beam shaping that generates lines, tubes or curved segments of focused light on distances that exceed the Gaussian Rayleigh range by several orders of magnitude. We review the benefits and applications of nondiffracting beams for laser micro- and nano-processing in the general context of materials processing with ultra-short pulses in the filamentation regime. We highlight applications on ultra-high aspect ratio nano-drilling and direct laser processing along curves.

Keywords: nondiffracting beams, Bessel beams, Accelerating beams, ultrafast laser materials processing

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1. Introduction

Ultrashort pulse lasers allow for materials processing at micro and sub-micrometric scales. Cutting, drilling, bulk or surface processing operations are possible with side damage in the sub-micron range. Ultrashort pulses deposit energy with ultra-high contrast between affected and non-affected zones. Ultrafast laser processing has entered in a novel development stage since high average power sources are available. While initially dedicated to niche markets and fast prototyping, ultrafast lasers now entered the markets of mass fabrication (solar technologies, consumer electronics, automotive) [1].

In this technological context, high-speed processing of large surfaces and deep drilling of materials are needed. A number of recent progress in terms of laser scanner technology and parallel processing have already greatly increased the throughput of laser processing. In this review, we show that using *non-diffracting beams* is an extremely powerful leverage to change the efficiency of the laser-matter interaction for high speed material processing and ablation.

Beam shaping has been developed and used for all types of laser processing techniques, mainly in 2D surface processing and shallow drilling of materials: the intensity distribution of the beam was optimized for one specific focusing plane to produce the desired mark (e.g. flat-top profile on a square or on a disk[2], arrays of spots[3] etc). In contrast, we will focus in this review on beam shapes optimized over long beam propagation distances.

Non-diffracting beams or diffraction-free beams are defined as propagation invariant solutions of the Helmholtz equation. Finite-energy realizations of these beams define extended focal lines, on distances much longer than the Rayleigh range. Several publications review works on laser processing with Bessel beams but separately from nonlinear optics of nondiffracting beams [4, 5, 6]. Here, we review the aspects of beam shaping and nonlinear optics that are directly relevant to ultrashort laser processing. In addition, we will review the extension to recently developed shaping techniques for the generation of "accelerating beams" that share many of the properties and benefits with nondiffracting Bessel beams.

We will first review the basics of the generation of nondiffracting beams, then the main important results of their nonlinear propagation in transparent media and their applications to laser processing, both in terms of surface processing and high aspect ratio deep drilling in transparent media. This will be performed in the general context of materials processing with ultrashort filaments, or more precisely, within elongated focal regions. The last section is dedicated to accelerating beams, which possess an extended focus that is curved in space. The typical example for accelerating beams, the *Airy beam*, is propagation-invariant *i.e.* diffraction-free, with a parabolic trajectory[7]. Other beams that exhibit an intensity peak propagating along arbitrary trajectories have been developed and have found applications in terms of laser processing of curved profiles. We will detail the generation of accelerating beams and its application to laser ablation of curved profiles.

2. Diffraction Free Bessel beams

Bessel beams. Bessel beams are produced by the interference between plane waves propagating along the surface of a cylindrically symmetric cone, i.e., having their k vector along a generatrix of the cone, crossing the optical axis at the same angle θ [8].

The interference produces a central lobe with high intensity, surrounded by circular lobes. McLeod showed in 1954 that Bessel beams can be generated from conical lenses, *i.e.* axicons (cf figure 1)[9]. It is only in 1987 that Durnin *et al* generated experimental beams exhibiting the diffraction-free property of Bessel beams, i.e. quasi propagation-invariant beams [10]. Indeed, the cylindrically symmetric field amplitude $E(r, z) = E_0 J_0(k \sin\theta r) e^{ik\cos\theta z}$ is a solution to the scalar Helmholtz equation. k is the modulus of the wavevector and θ is the angle made by the geometrical rays with the optical axis, which we will refer to as "cone angle".

This formal solution is of infinite energy. Experimentally, only apodized

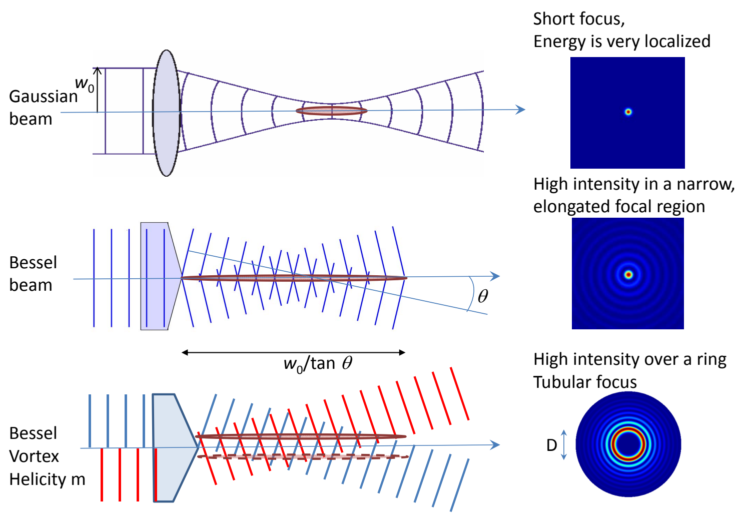

Figure 1: Schematic wavefronts and transverse intensity distribution for (top) Gaussian beam, (center) Bessel beam, (bottom) Bessel vortex beams

versions are physically realizable due to the necessary limitation in numerical aperture.

As an example, Bessel Gauss beams are obtained by sending a Bessel beam through an axicon and can be viewed as an ideal Bessel beam apodized by a large Gaussian beam. As the large Gaussian beam follows the laws for Gaussian optics, the Bessel-Gauss beam has invariant beam lobe positions the peak intensity and the number of visible lobes decrease far from the focus [11]. In the propagation direction, the full width at half maximum of the focal region for an apodized Bessel beam defines the Bessel zone. A Bessel-Gauss beam is produced by a Gaussian beam carrying a linear spatial phase $\Phi(r) = k\sin(\theta)r$ which reshapes the Gaussian beam into a Bessel profile. Within the stationary phase approximation of the Fresnel integral, the on-axis intensity profile is, in air : $I(r=0,z) = 4P_0kz\sin^2\theta e^{-2(z\sin\theta/w)^2}/w^2$, where P_0 and w are respectively the power and the waist of the input Gaussian beam [12, 13]. The spatial spectrum of a Bessel beam is a thin annulus of intensity centered at the mean transverse wavevector $k_{r0} = k\sin\theta$ with a FWHM width Δk_r inversely proportional to the

Gaussian beam waist w[13].

Therefore, close to the optical axis (*i.e.* over very few lobes) and in the scalar approximation, a typical Bessel beam radial amplitude distribution can be approximated by $E(r) = E_0 J_0(k \sin\theta r)$, where the peak amplitude E_0 can be determined by the stationary phase approximation. The polarization of Bessel beams is usually very close to the one of the input beam provided the cone angle is within the paraxial approximation. Detailed calculations and measurements have been reported since longitudinal polarization can be created with radially polarized beams[14, 15].

Bessel vortices are very similar to Bessel beams and are also diffraction-free solutions : they only differ by the fact that they carry an optical angular momentum, *i.e.* a vortex charge , and the main lobe in their intensity distribution is ring shaped as for high-order Bessel functions [16]. Details of the structure can be found in references [17, 18].

Figure 1 compares the structure of the geometrical rays and wavefronts propagating in a Gaussian, Bessel and Bessel vortex beams. In Bessel beams, all rays cross the optical axis with the same angle. In Bessel vortex beams, the rays have all the same angle with respect to the optical axis, but are tangent to a tube, forming a caustic [17, 18].

Experimental realization. A number of experimental techniques have been developed to produce Bessel beams, which can be split in two main families. "Direct" space shaping transforms a plane wave or its approximation into a Bessel beam by applying a linear ramp of phase with cylindrical symmetry. This can be performed with an axicon [9], sometimes processed at the exit side of an optical fiber [19, 20, 21]. More details on fiber-based Bessel beam shaping are reviewed in reference [4]. Other ways of shaping use holograms [22], spatial light modulators (SLM) [23, 24, 25] or diffractive optical elements [26, 27]. This approach allows for high-energy throughput, which is particularly important for applications in nonlinear optics or laser processing. When using axicon lenses, one experimental difficulty is the roundness of the tip, at the beam center. The roundness generates imperfections in the beam that can be corrected by further Fourier filtering [28, 29]. Approximation of Bessel beams can be generated with tunable acoustic gradient index of refraction lenses [30]. For applications, 4-f telescopes are used to generate Bessel beams with long working distance and in combination with Fourier filtering [31, 24, 25].

The second family of nondiffracting Bessel beam shaping is Fourier space shaping, where an *amplitude* modulation is performed. In the spatial frequencies, i.e. Fourier space, an ideal Bessel beam is a circle of amplitude $A(k_r) = \delta(k_r - k_{r0})$ [10]. Experimental realizations form an annulus of light in the back focal plane of a lens, and the Bessel beam is formed around the front focal plane, as the Fourier transformation. Beam shaping is based either on a hard aperture annular slit, or with Spatial Light Modulators or holograms, where only a thin annulus of light is formed in the first order of diffraction [10, 32, 22, 33, 34, 35, 8]. The longitudinal on-axis distribution of intensity in the Bessel beam $I(r = 0, z)$ depends on the exact distribution of the annular distribution of amplitude in the Fourier plane. For instance, a perfect flat-top ring distribution generates a Bessel beam with a longitudinal profile $I(r = 0, z) \sim (sin(az)/z)^2$ [33].

Other means include i/conical diffraction. The coupling between propagation direction and polarization in a biaxial crystal, has been recently used to produce Bessel beams [36]; and ii/ lasers designed to produce Bessel modes. The principle of operation is the use of a diffractive optical element as the end-cavity mirror[37].

Bessel vortex beams are produced either by applying a vortex to a Bessel beam, for instance by adding the vortex phase in the phase mask of the SLM, or by generating a Bessel beam from a Gaussian beam already carrying a vortex charge [22, 13, 17]. The vortex charge can be added with a vortex plate either in the Fourier or in the direct space [38, 39].

Spatio-temporal aspects. In the case of ultrashort pulses, the way of Bessel beams are generated is important because it determines the phase relation between the different temporal frequencies of the pulse. With refractive axicons, if dispersion is neglected over the pulse bandwidth, the spatial phase applied by refractive axicon is $\phi(r, \omega) = \frac{\omega}{c} r \sin\theta$. Using holograms the spatial phase applied is constant with frequency $\Phi(r) = k_0 r \sin\theta$ [40, 41]. The dependance of spatial phase on the temporal frequency impacts on the spatio-temporal shape of the Bessel pulse. In the first case, when spatial phase varies linearly with frequency, the generated Bessel pulse is called an *X-wave* because the amplitude distribution in the spatio-temporal spectrum (k_r, ω) has the shape of a X. The pulse is also X-shaped in (r, t) space. Importantly, the apparent speed of propagation of the on-axis lobe is $v = c/\cos\theta$ where c is the speed of light. The superluminal $(v > c)$ propagation speed has generated a great interest but is still physical because the photons all propagate at speed c. Superluminality can be interpreted as a "scissor effect" [40, 5, 42]. This behavior has been experimentally characterized in [43] by measuring the speed of the ionization front of a femtosecond pulse by shadowgraphy and in [44] by direct FROG measurement. It does have important applications in nonlinear optics since nonlinear index deformations can be generated with superluminal speeds [45].

In contrast, when the spatial phase is constant with temporal frequency, the Bessel beams generated are *Pulsed Bessel Beams*. The shape of the pulse is spatially a Bessel beam with a temporal Gaussian envelope. In this case, the virtual on-axis speed of the pulse is $v = c\cos\theta$ [46, 42], and is sub-luminal.

Generating Bessel pulses with Spatial Light Modulators is a specific case because the phase applied to the Gaussian laser beam by the liquid crystal array is not constant with the optical frequency. This phase is equal to the one applied by an axicon, but wrapped (most often over 2π). The spatio-temporal behavior is locally similar to Bessel-X pulses but can be approximated to Pulsed Bessel Beams over long propagation distance. It has been detailed in reference [47]. It is important to note that in all cases, the pulse duration of the input laser beam is preserved on the optical axis.

Modifications of the intensity patterns in Nondiffracting beams. For applications in microscopy, nonlinear optics or laser materials processing, it is sometimes important to modify the evolution of the on-axis intensity. Cizmar *et al* have developed a technique based on shaping the Fourier spectrum to engineer intensity profiles such as flat-top or linearly increasing ramps of intensity [33]. Ouadghiri-Idrissi *et al* have developed a similar approach that preseves the energy throughput [48]. In these approaches, the radius of the main intensity lobe of the Bessel beam is preserved but the relative intensities between the lateral lobes of the beam are modified.

A number of transverse profile variations of Bessel beams have been developed such as spiraling Bessel beams and helicon waves [49, 50] that can be viewed as accelerating beams [51]. Slight distortions of the input hologram allow for tilting and curving the beams along arbitrary trajectories [52]. Suppressing the side lobes of Bessel beams generates "needle beams". These beams have been generated by adapting the diameter of the axicon to generate only a single central lobe, thus performing an automatic apodization [53].

Self-reconstruction and resistance to aberrations. Bessel beams are "self-healing": the main central lobe can reconstruct after encountering an obstacle. This property arises from the conical flux of energy with the angle θ with respect to the optical axis. This has first been used for optical trapping with many particles [54]. The reconstruction distance after an obstacle of diameter a is $\Delta z = a/(2\cos\theta)$ [55, 53].

The spatial spectrum of a nondiffracting Bessel beam is highly peaked around the radial wavevector k_{r0}. This strongly reduces the impact of optical aberrations such as spherical aberration of a glass plate. Broky *et al* have investigated the resistance of Bessel beams to turbulent conditions such as those occurring in the atmosphere [56, 57].

The properties of spot-size invariance, resistance to obstacles and aberrations accelerated the interest for the use of nondiffracting beams in the fields of particle trapping [8, 58, 59] and light sheet microscopy [60, 61].

3. Laser surface processing with nondiffracting beams

The nondiffracting property of Bessel beams has been very early identified for laser processing. In contrast with Gaussian beams where the transverse and longitudinal dimensions of the focal zone are linked by a single parameter, the length of the Bessel zone and the main lobe transverse size can be independently adjusted by two free parameters: the initial Gaussian beam waist and the crossing angle θ determined by the axicon apex. Therefore, small spot size (down to $\lambda/2$, with λ the laser central wavelength) can be achieved over arbitrary propagation distances.

This offers the possibility of maintaining identical illumination conditions on a workpiece whatever its position within the Bessel zone. This requires however that only the central lobe is involved in the process.

In terms of surface processing, this has opened three main applications, not necessarily implying ultrashort pulses: i/ laser processing of curved surfaces [62] ; ii/ laser processing of biological material where the position of the sample is not well known [63, 20] and iii/ ultrafast laser processing with non-critical positioning [25]. Ultrashort pulses can induce optical breakdown and ablation down to nanometric transverse dimensions, but in return, this requires nanometric absolute positioning of the workpiece versus the Gaussian focal spot [64, 65]. Nondiffracting beams solve this issue and were used to drill sub-micron holes in glass, graphene and metallic films [25, 66, 67]. We note that surface processing with nondiffracting beams is more beneficial for films or materials that are opaque at the laser wavelength. In transparent materials, the long depth of focus of Bessel beams can induce a damage or index modification well below the surface of the material, as it will be described in section 4.

Reference [68] reported Bessel beam formation with annular slits and their application to surface ablation over subwavelength diameter. Nondiffracting beams with modified shapes or vortices allowed the generation of micro and nano-disks on graphene films [69] and the investigation of ablation threshold and the ablation probability [70, 71]. Cheng and Polynkin reported annular surface crater formation with ultrashort pulse ablation of fused silica with high order Bessel beams [72].

Combination of lens and axicon focusing allows for reducing the Bessel zone over which laser pulse energy is spread. It allows for preserving the Bessel profile and enhances the local intensity on the central lobe: this approach has been used for silicon micro-drilling [73].

4. High aspect ratio ultrafast laser processing with nondiffracting beams

High aspect ratio laser processing. Deep drilling in materials is an issue for many technological fields. The aspect ratio is a representative figure of merit of the technical difficulty for drilling: it is the ratio of the channel depth over the channel diameter. While aspect ratios up to 10:1 are common whatever the drilling or cutting technique (laser, mechanical drill, water jet, DRIE, Focused Ion beam.. etc), it is much more difficult to extract matter out of channels with high aspect ratio exceeding $\sim 10:1$. This is even more difficult for channels in sub-micron diameter range.

Nondiffracting Bessel beams and needle beams are *a priori* potential candidates for high aspect ratio drilling because of the ultra-long focal distance. However, it is important to note that drilling with Bessel beams is only possible in transparent materials. Indeed, in opaque materials, the conical flux of energy generating the central lobe cannot propagate. Bessel beam drilling of metals generates V-shaped craters where the opening angle is close to the Bessel beam angle [74]. Therefore, in the following, our interest will be only for transparent materials, unless explicitely mentioned.

Transparent materials processing by ultrashort pulses. Ultrafast lasers are well suited to processing transparent materials because they can deposit energy at the surface or in the material bulk with an extreme degree of confinement. In brief, ultrashort laser material modification process is the following. High intensities (typ. 10^{13} W.cm^{-2}) reached in ultrashort pulses allow multiphoton ionization to generate free-electrons in the conduction band. The transition involves typically between 2 and 15 photons depending on the bandgap and illumination wavelength [75, 76], where the number of photons is determined by the ratio between the energy to promote an electron from the valence band to the conduction band and the photon energy. The plasma of free-electrons and holes can further absorb light through collisions. The hot electron gas then relaxes by carrier-phonon collisions and transfers its energy to the initially cool lattice. Phase transformations, diffusion, hydrodynamic effects occur on timescales from several picoseconds to microseconds. The full description of laser-matter interaction and relaxation processes is out of the scope of this review and more details can be found in references [77, 78, 79, 80].

Nonlinear propagation in transparent materials. It is important to realize that the propagation of light into the medium is nonlinear and highly impacts on material ablation or modification process. The nonlinear propagation of Gaussian beam pulse is difficult to predict, even in the simplest case of the propagation in defect-free media (*i.e.* single shot damage). Three main sources of nonlinearities are : i/ the nonlinear Kerr effect that modifies the index of refraction of the medium n_0 as $n = n_0 + n_2 I$, with I the local intensity of the field , ii/nonlinear ionization, which rate can be described by Keldysh formulation as a first approximation [81] and iii/ the free-electron plasma. The latter induces defocusing since, in first approximation, it induces a refractive index change of $\Delta n = -\frac{\rho(\mathbf{r},t)}{2\rho_c}$ where $\rho(\mathbf{r},t)$ is the local density of free electrons and ρ_c the critical plasma density at the laser central wavelength ($\rho_c \sim 1.7 \times 10^{21}$ cm^{-3} at 800 nm) [82]. The free-electron plasma also absorbs the laser pulse. Losses are typically modelled through the plasma Drude model [81].

These physical effects also greatly affect the pulse in time and spectrum through spectral broadening, self-steepening etc [82]. The previous list is not exhaustive : several other effects might need to be taken into account for relatively long propagation distances ($> cm$) or extremely short pulses (typically < 20 fs) : group velocity dispersion, Raman contributions to nonlinearities, etc.

Depending on actual intensities, saturations of the nonlinear effects do occur.

At moderate intensities and focusing (focal spot diameter $\sim 10\mu$m), it is relevant to describe pulse propagation and laser-matter interaction by two coupled equations. The pulse amplitude is described by the nonlinear Schrödinger equation [81, 83] describing the evolution of the envelope of the electric field, that is coupled to a rate equation describing ionization processes (multiphoton and tunnel ionization described by Keldysh formulation and impact ionization). Additional saturation terms taking into account the finite number of electrons in the valence band were described in reference [84] and the influence of self-trapped excitons was also modelled by several groups [77, 85].

We note that modelling laser-matter interaction in dielectrics is still a hot topic because of the technical difficulty to take into account the numerous effects of out-of equilibrium physics, with large gradients [80, 86, 87, 88]. Tractable numerical models require numerous approximations on plasma dynamics, which strongly restrict the domain of validity of the models. Linking pulse propagation to the damage is an even higher challenge [80, 85].

In conclusion of this section, the nonlinear propagation of Gaussian pulses in transparent materials at high intensities yields highly distorted pulses, both in space and time.

Filamentation. Under the combined action of Kerr self-focusing and plasma defocusing, the nonlinear propagation of Gaussian pulses typically tends to a dynamical regime of light propagation where one or several hot-spots self-form in the beam over extended propagation distances, leaving long tracks of plasma in their wake. These structures are called filaments [89, 90, 82].

The extended propagation length exceeds by several orders of magnitude the Rayleigh range corresponding to the diameter of the hot spots. This effect occurs both in solids and gases and the self-healing properties of filaments rendered them particularly interesting for atmospheric applications [91].

In this research direction, the formation of elongated damages by filaments were attractive for laser materials processing at high aspect ratio.

High aspect ratio processing by filaments. High aspect ratio damages induced in by filaments in transparents solids were reported by several groups [92, 81, 93, 94, 95, 96]. The process has been also investigated by means of pump-probe analysis of the transient dynamics of free-carrier generation and material damage [97]. It has been observed that the filamentation process is enhanced by spherical aberration, such as the aberration induced by propagation through thick samples (rear side focusing conditions). Nanochannels with aspect ratio $\sim 20:1$ were generated by single shot focusing on rear side of glass [98]. Elongated channels were obtained in single shot within the bulk of fused silica and PMMA [99, 100, 101, 102]. More recently, Herbstman and Hunt created high aspect ratio nanochannels in single shot focusing at the exit side of fused silica plates[103]. Ahmed *et al* reported that the spherical aberration induced by thick glass plates inserted between the focusing lens and the sample allow drilling extremely long voids in glass in single shot [104].

A glass cutting technique has been developed, where filamentation tracks create elongated damages in glass: creating those tracks next to each other, at translation speed of the workpiece on the order of 1 m/s, guides mechanical cleaving or self-cleaving, depending on the type of glass and illumination conditions [102].

However, the structure of the filament in this case is bounded to the degree of control of the competition between Kerr effect and plasma defocusing. The complex ionization dynamics of glasses makes this technique highly difficult and sensitive to input conditions (beam diameter, beam quality, pulse duration,energy, position of the sample, etc).

Nonlinear propagation of nondiffracting beams. Nondiffracting Bessel beams are advantageous over Gaussian beams for laser bulk processing because their nonlinear propagation permits a stationary regime with negligible spatio-temporal dynamics and generates a uniform plasma track. This regime is stabilized by high nonlinear losses and allows for a high degree of control of light-plasma interaction in the bulk of the material with reduced spatio-temporal distorsions.

Nonlinear propagation of nondiffracting Bessel beams has generated an intense interest when it was realized that filamentation of Gaussian beams spontaneously generates structures that are highly similar to Bessel beams: conical waves and self-healing properties [105, 106, 107, 108, 109, 110].

Polesana *et al* identified three propagation regimes for intense Bessel beams [111]: i/"weakly nonlinear", where losses are negligible and is featured by oscillating peak intensity and transverse lobe compression [112]; ii/ "unsteady Bessel filamentation", characterized by high peak intensities yielding periodic optical breakdown and damage [113] and where the pulse propagation shows spatio-temporal dynamics; iii/ "steady Bessel filamentation" shows an absence of dynamics. In the latter regime, high losses occuring in the central lobe are compensated by the energy flow from the surrounding lobes (conical flux, playing the role of a reservoir).

Propagation invariant and monochromatic solutions to the nonlinear Schrödinger equation exist [114]. In comparison to the Bessel beams, they are featured by lobe compression reflecting the optical Kerr effect and to a power flux from the periphery toward the main lobe where nonlinear losses are localized [115]. The low intensity tail of the beam can be viewed as a superposition of two cylindrical Hankel waves with unequal amplitudes. Nonlinear losses in the intense part correspond to the difference between the power flux for the inward and the outward Hankel waves. This feature is at the origin of the name of Nonlinear Unbalanced Bessel Beams (NLUBBs) given to these solutions. Like Bessel beams, NLUBBs have apodized counterparts carrying finite energy, which play the role of attractors in the filamentation dynamics of Gaussian beams. NLUBBs with similar features can be found whatever the type of nonlinear losses or nonlinear refraction index.

The stationary, or steady, regime of Bessel filamentation was investigated in the context of applications that require the generation of a uniform plasma channel in air or in liquids [116, 117, 118]. Due to the conical structure of Bessel beams, the photons creating the plasma in the central lobe have travelled along trajectory crossing regions only with low intensity, with moderate nonlinearity and losses. This allows minimizing the effect of plasma defocusing and optimizing localization of light as energy arrives from the side in the focal volume without crossing the plasma [119].

Experimentally, the choice of input conditions impacts on the dynamics of Bessel filaments. Polesana *et al* have shown that the progressive intensity rise and Bessel beam formation in the nonlinear medium yield stationary filamentation regime. In contrast, when the Bessel beam is formed before the linear/nonlinear interface, it can lead to the dynamical unsteady filamentation regime [120].

Laser processing of transparent materials with Bessel beams. Marcinkevicius *et al* reported the first optical damage by Bessel beams in glass with ultrashort pulses [31]. After multiple shot illumination of glass, straight modifications of the index of refraction were observed in fused silica. J. Amako *et al* realized similar marking in glass with Bessel beams, followed by selective chemical etching to process high-aspect ratio channels [26]. Grooves in fused silica were also processed with a similar approach [121]. In the multishot regime, the progressive build-up of a damaged zone can be interpreted as a spatially nonlinear absorption : the stationary regime is favorized by the stronger absorption of defect centers within the central lobe. The latter regime has also been used to write volume Bragg gratings in fused silica [122].

Bhuyan *et al* produced the first application in the ablation regime in single shot [123]. A key element is the high cone angle used: θ was varied between 16 to 26 degrees in air, corresponding to 11 to 17 degrees in glass. Nanochannels could be drilled in glass with aspect ratio up to 100:1. At that time, no other technological mean could reach so high aspect ratio in short processing time.

Figure 3 summarizes the drilling mechanism at play. During the propagation of the laser pulse in the dielectric, a plasma of electron-hole pairs is created by the Bessel beam. High cone angles, exceeding $15°$ play a key role in maintaining a stationary regime even at high intensities and in building a near uniform plasma density. Lower cone angles do not always provide a stationary regime.

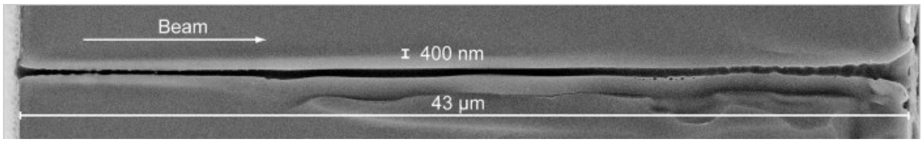

Figure 2: Proof of principle machining of a through channel using Bessel beam conical half angle $\theta = 11°$ in glass and a 43μm thick glass membrane at single shot pulse energy of 3.1μJ. Reprinted with permission from reference [123]

At cone angles $\theta \sim 7°$ in glass, transitions to unsteady Bessel filamentation regime were observed when the Bessel zone crosses the sample entrance surface [124]. A novel technique of chirped pulse spectral transmission confirmed the stationary behavior of high-angle Bessel beams at ablation-level conditions [125].

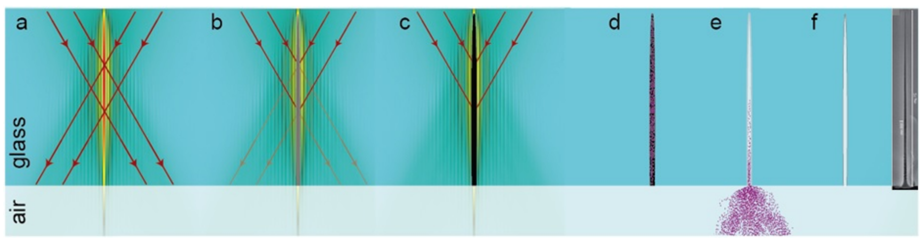

Figure 3: Physical sequence of drilling. (a) Bessel beam propagation; (b) creation of a plasma channel of electron-hole pairs; (c) plasma is highly absorbing and creates avalanche; (d) energy transfer from the electron-hole plasma to the lattice; (e) phase change and material expellation and/or material compaction; (f) cooling and an elongated void is left.

After the build-up of the plasma of electron-hole pairs, the hot electron gas transfers it energy to the cool lattice, which undergoes rapid phase change (melting, evaporation, atomization). Two mechanisms for channel opening are at play : i/ material compression such as in the case of formation of voids in silica and sapphire[126]; ii/material expellation through the channel itself.

In the case of nanochannel drilling in Corning 0211 glass [123], material removal is the main mechanism at play. It was observed that drilling occured only when the Bessel zone was crossing the exit side. Focusing in the bulk of the material led to elongated index modifications with no void. In contrast, another study with Bessel beams with picosecond pulse duration, elongated voids could be realized completely in the bulk of fused silica , suggesting the second mechanism [127]. The influence of cone angle, thermal diffusivity coefficient of the material (linked to rapid or fast cooling from liquid to solid phase), cooling and ejection dynamics of the plasma column are potential parameters influencing the difference in behaviour. Understanding the thermomechanical evolution of the material after laser energy deposition remains a challenging problem calling for further investigations.

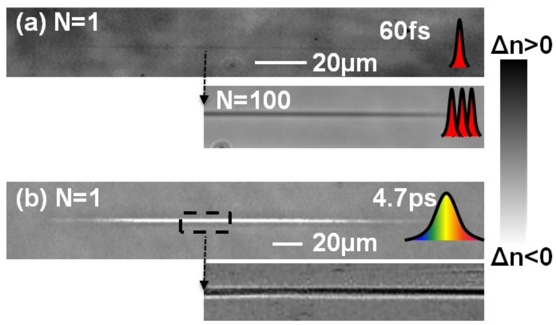

Figure 4: Phase constrast microscopy images of Bessel beam induced high-aspect-ratio structures in fused silica at moderate focusing conditions (theta = 8.3). (a) Single-shot positive refractive index structure at 1.72 μJ pulse energy and 60 fs. (inset) Multipulse (N=100) smooth refractive index structure for 1 μJ, 60 fs pulse irradiation conditions. (b) Uniform void structure at 7 μJ/pulse, 4.7 ps. (inset) FIB/SEM Cross sectional profile of the fabricated structure at ps pulse durations

The influence of input pulse duration has been investigated [127, 128]. It was reported that picosecond pulse durations led to enhanced damage in fused silica. Figure 4 shows that moderate focusing of Bessel beams with variable pulse durations can tune positive and negative index changes. Indirect measurements of free-carriers density in the plasma indicate the higher absorptivity of the plasma created by picosecond pulse durations [129]. However, microchannel drilling in borosilicate glass was reported with aspect ratio 1200:1 for 50 fs pulse duration[130]. The interpretation of the differences between picosecond and femtosecond regimes is nontrivial because at high degrees of ionization, a number of different mechanisms change light absorption : bandgap shrinking, ultrafast phase change during the picosecond pulse, dynamics of trapped states, etc. [86, 131, 85, 132].

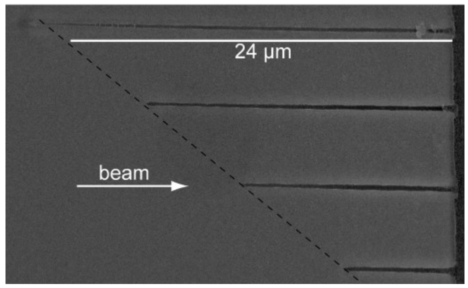

Figure 5: SEM image showing four machined channels with sample displacement differing by 4μm in the horizontal direction. Reprinted with permission from reference [123].

In contrast with Gaussian beam filamentation, the longitudinal position of the channels is highly predictable, as shown in figure 5 [123]. These results highly impact on the field of glass cutting. Single shot writing of channels in lines generates a stress in glass that can be used to cleave the material at speeds on the order of 1 m/s. Stress generation by Bessel beams in glass and kerf-free cutting of glass have been investigated by Tsai *et al* [133] in the femtosecond regime. High speed cutting of glass was reported using picosecond regime with a slightly larger kerf [128]. Processing of silicon with Bessel beams was investigated in the transparency window at laser wavelength of 1.3 μm [134]. It was reported the absence of damage in the bulk, which was attributed to nonlinear losses before light reaches the focus.

5. Tubular Plasmas

It is attractive to drill large diameter holes (several tens of microns) with annular beams, such as those produced by optical vortices. However, during nonlinear propagation of annular beams in dielectrics, modulation instability tends to break-up the ring of intense light into several hot spots [135, 136, 137].

Recent work from several groups has shown that Bessel beams carrying vortex charge, *i.e.* "Bessel vortices" can also sustain stationary propagation.

Monochromatic and propagation invariant nonlinear solutions of the nonlinear Schrödinger equation in form of a Bessel function carrying angular momentum has been found [138, 139]. Experimental and numerical investigations of femtosecond Bessel vortices in fused silica have shown the transition between three regimes when the pulse energy was increased. The stationary regime creates a tube of plasma in glass or in air following the main lobe of intensity with obvious applications to laser materials processing [140, 141]. This regime is sustained over a finite range of energies. This energy range is extended when the cone angle is increased, as for the case of zero-order Bessel beams. At higher intensities, nonlinear wave mixing generates supplementary spatial frequencies. First, the main intensity lobe breaks-up into several hot spots spiraling around the optical axis. This regime is close to what was observed in water by Shiffler *et al* [142]. At much higher intensities, in "speckle-like" propagation regime, multiple hot spots appear and disappear along the propagation distance. This regime is unexploitable for laser processing while is it still reproducible from shot to shot.

Proof of principle tubular index modification has been reported in glass from single shot Bessel vortices in the stationary regime [141, 143]. Picosecond laser processing with vortices has been investigated in single and multiple shot regime by Jedrkiewicz *et al* [144]. Altough tubular modification could be reported (see figure 6), drilling of cylindrical holes remains difficult because of the distorsions due to the roughness appearing after a few laser shots. Energy deposition is not efficient enough to realize cylindrical voids. Novel strategies are needed for this aim.

6. Accelerating beams and laser processing of curved profiles

The technological field of the fabrication of screens, smartphones, flat panels, microelectronics requires profiling of glass with rounded edges. Brittle materials with rounded edges are less prone to fracture than those cut with hard edges. In this section, we show that another family of beams, "accelerating beams",

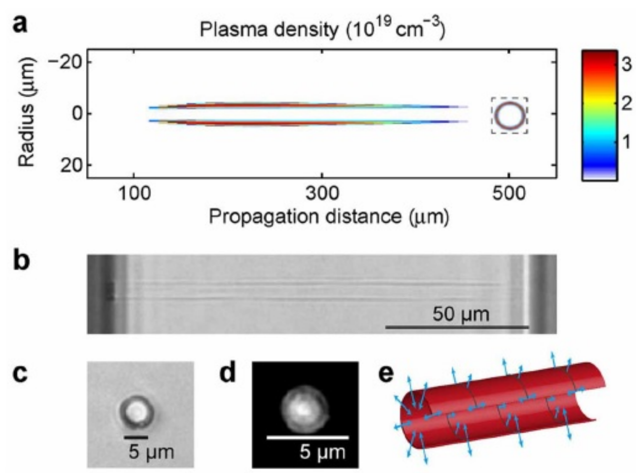

Figure 6: Tubular plasma generated in the stationary regime. (a) Tubular plasma density distribution at a pulse energy of 5 μJ. The inset shows the cross section of the plasma distribution at a distance of 300 μm (b) longitudinal view under transmission optical microscopy of a tubular damage produced in glass by single shot, high energy, stationary Bessel vortex, pulse energy 35 μJ and pulse duration 1 ps. The damage extends from one sample side to the other. (c) Transverse section of the damage observed in identical conditions the same beam at pulse energy of 20 μJ. The bright central region shows high index modification of the core of the tubular region. (d) Image of near-field output guided light in the structure shown in (c), at an input wavelength of 632 nm. (e) Schematic view of the propagation direction of mechanical and thermal waves expanding outward and inward (arrows) from the excited tubular sheet volume (circle). Reprinted with permission from reference[141].

provides interesting pathways to solve this issue.

Currently, after laser cutting, glass panels are mechanically polished to reach rounded edges. This is time consuming and not useable for thin glass panels. Glass with bull nose profile can be obtained by straight cutting of chamfers (+45 and -45°). Mathis *et al* developed an approach for direct curved laser profiling with arbitrary profiles[145]. In this section, we will review recent results obtained with accelerating beams whose main intensity lobe propagates along an arbitrarily curved trajectory. The beam shaping technique for generating accelerating beams conserves the pulse duration on the trajectory, and therefore allows for ultrafast laser processing. We will review results in terms of laser curved profiling and curved cutting.

Accelerating beams. In 2007, Siviloglou and Christodoulides identified the analogy between a nonspreading quantum wavepacket [146] and an optical field propagating along a parabola [7, 147]. The Airy beam is a solution of the paraxial wave propagation equation. Its transverse intensity profile is invariant along the propagation, while the position of its maximum follows a parabola. As Bessel beams, Airy beams are of infinite energy but apodized versions have been well identified [7]. These results have triggered an intense interest. Applications have been developed in almost all fields of optics and photonics [148]: optical trapping [149], microscopy [150], nonlinear optics [151, 152, 153], plasmonics [154] and even electron waves [155].

Two important results for the field of laser processing are that, like Bessel beams, Airy beams are self-healing [56] and Airy beams sustain stationary nonlinear propagation [156]. As for nonlinear Bessel beams, stationarity is supported by an energy flux from the low intensity tail of the beam toward the intense main lobe where nonlinear absorption occurs. Curved plasma generation in air with paraxial Airy femtosecond pulses has been reported in 2009 [157].

Experimentally, Airy beams are generated by applying spatially cubic phase in the focal plane of a lens, realizing an optical Fourier transform. However, for applications to laser processing of curved profiles, Airy beams do not show enough bending on short distances. Froehly *et al* [158] and Courvoisier *et al* [159] have therefore developed an approach to shape non-paraxial beams. This has been performed by identifying that Airy beams can be interpreted in terms of optical caustics, *i.e.* a family of optical rays tangent to a curved trajectory [160]. In the framework of optical catastrophes, the transverse Airy function arise from a fold catastrophe (see supplementary information in reference [161]).

From these observations, shaping along arbitrary convex trajectories could be realized experimentally by applying a spatial phase to a Gaussian beam [161, 158].

Accelerating beams cover the family of beams having a main intensity lobe following a curved trajectory. The transverse profiles are not necessarily diffraction-free. In 2D, the FWHM of the main intensity lobe of an accelerating beam is linked to the local radius of curvature of the trajectory: $\Delta x = 1.630[R/2k^2]^{1/3}$, where k is the wavevector and R the local radius of curvature of the trajectory [159]. The transverse lobe dimension therefore evolves: higher focusing is reached for highly curved trajectories.

This work has been further extended in highly non-paraxial regime (numerical aperture exceeding 0.4) with Fourier-space shaping. In this case, experimental realization requires a correct description of the correspondance between the optical frequencies and the position in the back focal plane of high numerical aperture microscope objectives. Arbitrary nonparaxial trajectories (circular, parabolic, quadratic) were experimentally generated with bending angles filling the full numerical aperture of a microscope objective of NA 0.8 [162]. Simultaneously, analytical derivation from Maxwell equations predicted the same results [163, 148] and was extended since then to many other geometries and properties [52].

As for the case of Bessel beams, the spatio-temporal properties of Airy beams depend on the space and mean of shaping. Direct space shaping [159] and Fourier space shaping [164] were investigated and showed that in both case, pulse duration is preserved on the caustic trajectory. In the first case, the pulse

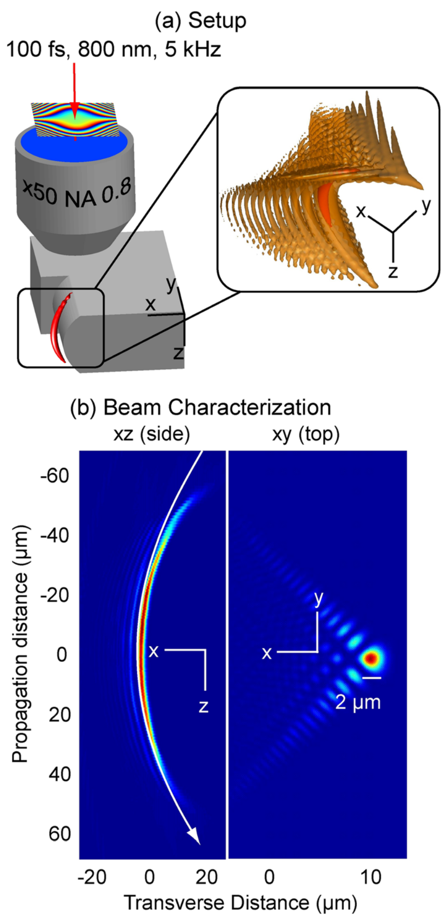

Figure 7: (a) Concept of laser processing of curved profiles with acccelerating beams. (b) Intensity distribution of the accelerating beam in the longitudinal xz plane and the transverse xy plane. Reprinted with permission from reference [145].

travels along the curved trajectory while in the second, the whole trajectory is illuminated simultaneously.

Experimentally, accelerating beams are generated with spatial light modulators, diffractive optical elements or phase plates produced in fused silica [165].

Laser processing of curved profiles. Early demonstration of curved damage in glass induced by an accelerating femtosecond pulse was reported by Froehly *et al* [158]. Mathis *et al* investigated two approaches of materials processing with accelerating beams: side processing of edges and direct curved trench machining[145]. In both cases, the beam used was obtained by interference of two 1D accelerating beam profiles, locally generating an intensity pattern of $I(x, y) \sim |Ai(x/w_0)Ai(y/w_0)|^2$. A main intensity lobe propagates along predefined curved trajectory. This allows reaching higher peak intensities and avoiding break-up of 1D profiles into hot spots by modulation instability.

It is however important to note that only a part of the input pulse energy contributes to build the main intensity lobe along the propagation, in contrast with Bessel beams. As shown by Kaganovsky *et al*, only a part of the optical rays in a 2D Airy beam formed by such interference cross the central lobe [166].

Curved edge processing of silicon and diamond were reported. This is performed by back and forth translation of the beam along the edge with progressive penetration toward the central part of the workpiece, as shown in the concept figure 7 adapted from reference [145]. With this approach, arbitrary profiles could be imprinted on the material with excellent agreement with expected profiles. The limitation of this approach is the formation of diffusing roughnesses on the edge of the structure. Depending on illumination conditions, the roughnesses modify the local ablation rate and discrepancies with expected profile appear.

The second approach developed in this work was detailed in reference [167]: direct translation of the accelerating beam into an opaque material such as silicon for a femtosecond laser centered at 800 nm, opens a curved trench. In this case, the intensity distribution on the top surface of the workpiece is an essential parameter. Indeed, the opening of the trench let the light penetrate into the medium and follow the curvature of the beam trajectory.

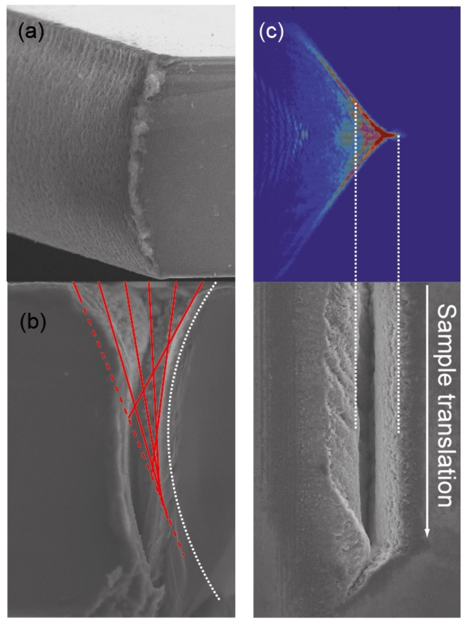

Figure 8: (a) Curved processing of a silicon wafer of thickness 100 μm. (b) direct curved trench machining of silicon. The geometrical rays show where the drilling stops following the curved trajectory. (c) Intensity distribution on the top surface of the silicon sample : the trench opening is performed by the two side lobes of the beam.

In principle, this approach can be applied to transparent materials. However, refraction at the air/dielectric interface reduces the effective curvature of the accelerating beam inside the material. The multishot drilling process is then expected to become more complex.

Accelerating beams have also found another application in the field of laser processing. *Abruptly autofocusing waves*, produced by the cylindrically-symmetric interference of Airy beams, produce an intense focus over a propagation distance shorter than the equivalent Rayleigh range [168, 169]. This property has been used to create localized ablation at the exit surface of transparent material, without damage in the region preceding the ablation zone [170].

7. Unsolved problems and future directions

Nondiffracting beams have open up a very novel way to create intense interaction over extended propagation distances. Particularly, the physics at play during channel formation is only partially uncovered. Pump-probe imaging of the plasma densities generated by Bessel beams have been realized [129], but a key difficulty of the analysis of the experiments lies in the extremely high gradients of plasma density generated over subwavelength transverse dimensions. Future work include the investigation of the exact influence of cone angle, thermal diffusivity and the hydrodynamics of the plasma column.

The investigation of the compression mechanisms and plasma/matter ejection during the formation of the nanochannels is an important task to explain channel formation and the different regimes of channel fragmentation oberved by Bhuyan *et al* [127]. This understanding is also essential to extend single shot nanochannel drilling to other transverse geometries such as channel drilling with vortex beams to obtain tubular channels or with curved accelerating beams. A key aspect is to control the *energy density* deposited during the laser-matter interaction.

Conclusion

The recent research developments on nondiffracting Bessel and accelerating beams open up new possibilities for fast processing of glasses. Nonlinear beam propagation in transparent dielectrics supports stationary regimes of Bessel and Bessel vortex filamentation. These regimes have generated unique pathways to control high aspect ratio ultrafast laser structuring and cutting. In contrast with Gaussian beams, the elongated focal line of nondiffracting beams allows for fast surface structuring at nanometric diameters without critical positioning of the beam on the workpiece. Thanks to their higher stability at high intensity, Bessel beams provide means to circumvent beam distortions that originate from nonlinear Kerr effect, ionization and defocusing by free carriers. This is of prior importance to control bulk processing of transparent materials.

The properties of accelerating beams are close to those of Bessel beams and very novel approaches for material separation and edge processing have been developed. These results have already attracted a wide interest in optics and photonics as well as in technological fields such as glass processing market. We anticipate that demanding applications will foster this research into solving those novel challenges.

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