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Tailored-Beam Ultrashort Laser Pulses

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Abstract: Laser pulses with picosecond and femtosecond durations are now indispensable tools in many scientific disciplines. These ultrashort pulses provide unique temporal resolutions for investigations of phenomena taking place in comparable time ranges. In addition, optical energies squeezed into such short time windows yield extremely high peak powers and intensities, paving the way to generate unprecedented light-matter interactions. On the other side, recent advances in spatial light intensity and phase modulation devices enables generation of laser beam profiles with intriguing physical properties. By merging the two realms, optics researchers can now have the ability to harness light both in space and time. These possibilities bring about new physical interactions and significantly facilitate many applications of lasers.

Keywords: Tailored-Beam; Laser Pulses

1. Introduction

Obtaining electromagnetic fields tailored to fit a particular purpose is a general goal behind most Optics research. In this regard, lasers are the most suitable sources, as they allow a broad control over temporal and spatial properties of light, either through a direct modification of the laser cavities, or by external modulation. Ultrashort laser pulses, with durations of picoseconds and femtoseconds are particular examples of temporal tailoring. Such pulses are generated through mode-locking of multi-longitudinalmode lasers [1]. These short pulse durations appeal scientists since they provide unique tools to investigate ultrafast phenomena happening in the same time scales [2]. The typical implementation for high temporal resolution experiments is the pump-probe setups, where the sample is excited by a short pulse, and thus caused alterations in the samples are probed by a second short pulse, which is controllably delayed with respect to the first. Another principle appeal of ultrashort lasers stems from the extreme high peak powers and intensities resulting from short durations. Indeed, these pulses provide highest localized electric fields available in controllable fashion in a laboratory. Such field magnitudes enable unprecedented laser matter interactions and nonlinear optical processes. Laser-ignited nuclear fusion [3], lightning trigerring [4,5], generation of extreme ultraviolet [6,7,?] or even gamma ray pulses [9],

probing the limits of quantum electrodynamics [10] are only a few examples of applications under current research efforts.

Laser light can also be tailored spatially. Laser cavities can support stable oscillation of certain transverse electric field profiles. Stable transverse modes are generally described by Hermite-Gauss or Laguerre-Gauss polynomials, in cartesian and cylindrical symmetries, respectively [11]. These polynomials provide a large variety by themselves. However, by using systems which can modulate the intensity and phase of light out of lasers, much more flexible beam shaping can be achieved. Laser beam shaping is experiencing an increasing interest from light sciences, especially due to recent improvements in optical manufacturing and computer-controlled light modulators. Through beam shaping, one can investigate the nature and propagation of structured light in a fundamental physical level; or one can also strongly improve or facilitate a creation application of light.

In this review, we summarize recent studies involving both temporally and spatially tailored laser light. We mainly focus on fundamental and applied studies involving ulrashort laser pulse, with specially shaped beam profiles. In the first part, we summarize the most commonly used beam profiles and beam shaping methods; and in the second part, we elucidate on selected applications.

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We should point out that this field is gaining increasing attention and it is impossible to list all approaches. Due to the nature of beam shaping, there are unlimited possibilities. Our intention is not to compile a complete list, but rather to draw attention to some of the recent and most dramatic results.

2. Commonly used beam classes

In this section, we summarize some commonly used beam profiles. In each case, we briefly layout the physical foundation and then explain methods for experimental realization.

2.1. Diffraction-free beams

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Diffraction is a result of wave nature of light. Diffraction phenomena appears when electromagnetic waves encounter finite apertures. Laser cavities involve small-aperture mirrors and lenses, hence they support stable oscillation of light only within a narrow angular spectrum. The eigensolutions of paraxial wave equation are the so-called Gaussian beams [11]. The propagation of these beams are self-similar, governed by diffraction. In particular, as the laser light propagates, Gaussian beams keep their profiles but scaled to different beam sizes. Diffraction also plays the determining role in laser beam focusing. The smallest focused beam size is limited by the laser wavelength (called the diffraction limit). The beam also stays focused within a narrow propagation distance (The Rayleigh range), proportional to the square of the beam size. The Rayleigh range also determines the region over which the intensity stays high, or the useful depth of focus.

The diffraction-free beams eliminate most of the restrictions above. In general, the beam sizes or profiles of diffractionfree beams does not change by propagation, and as a consequence, their intensity does not drop. The term refers to an idealization and real diffraction-free beams are not physical as they require infinite energy. Nevertheless, as explained below, experimental approaches can yield results not far from to the ideal cases and bring about significant advantages in applications.

2.1.1. Bessel beams

The notion of diffraction-free beams was first put forward by Durnin et al. [12,13]. They point out that the zeroth order Bessel function of the first kind is an exact solution of the wave equation in free-space, and this particular solution reproduces same intensity for all propagation distances [13]. The corresponding field profiles have simple mathematical description

$$E(\mathbf{r},t) = \exp\left[i\left(\beta z - \omega t\right)\right] J_0(k_r r) \tag{1}$$

In Eq. 1, r is the radial coordinate, k_r is the transverse wavevector and J_0 is the zeroth order Bessel function of the first kind. Note the the well-known plane wave solution is a spacial case for $k_r = 0$. While Eq. 1 provides a theoretical non-diffractive field, since this function is not square integrable, it corresponds to infinite energy, and hence is unphysical. On the other hand, physical solutions can be obtained by imposing a finite aperture function, and thus limiting the energy to finite values. Such finite aperture Bessel-beams are not perfectly diffraction-free, yet they are realistic and they provide quasi-diffraction-free propagation over a finite propagation distance. This behavior is a dramatic change as compared to Gaussian beams. Bessel beams with beam diameters close to the wavelength-limit can keep high intensities over orders of magnitude larger distances as compared to Rayleigh distances of Gaussian beams (of the same beam size).

Apart-from diffraction-free propagation, Bessel beams show another remarkable property, known as "self- reconstruction" (also referred to as self-healing). When the beam is partially blocked by an obstacle anywhere on the transverse plane, the beam shape can be reconstructed almost perfectly, after a brief propagation [14]. The Bessel beam pattern is formed by interference of the input plane-wave or Gaussian beam fromed into a conical surface. When part of the beam is blocked at a certain longitudinal plane, pattern at a later plane is unaffected (except for small diffraction effects) as it is formed by a different section of the input cone. The self-reconstruction property becomes useful for non-homogeneous and scattering media, as discussed in applications part below.

Experimental realization of Bessel beams has several alternative approaches. The most straightforward approach is to pass a Gaussian laser beam or large (plane wave-like) beam through a circular aperture, followed by a lens (for optical Fourier transform) [12]. A more energy-efficient method is to pass the beam through an axicon (i.e. conical lens) [15, 16]. The radial spatial phase imposed on a Gaussian beam transforms the light into Bessel beams in the near field. The resulting intensity profile is approximately given by [17]



Figure 1 Theoretical intensity profiles of a Bessel beam, generated by passing a Gaussian beam through an axicon. Top: 2-Dimensional transverse distribution. Center: Radial distribution. Bottom: Longitudinal on-axis distribution. In the calculations, light wavelength is taken as 633nm, input Gaussian beam size is $w_0 = 2mm$ and the base angle of the axicon 25-degrees. Axicon material is fused silica.

$$I(r,z) = \frac{4Pkr\sin\beta}{w_0} \frac{z}{z_{max}} J_0^2 \left(kr\sin\beta\right) \exp\left(-\frac{2z^2}{z_{max}^2}\right)$$
(2)

where P is the total power in the beam, w_0 is the input Gaussian beam spot size at waist, k is the wavevector magnitude, β is the half cone angle of the beam after the axicon, z is the longitudinal position and z_{max} is the position at which maximum axial intensity occurs. Radial and longitudinal intensity cross-sections, as well as a two-dimensional transverse profile of a typical Bessel beam, calculated from Eq. 2 is given in the Fig. 1 below.

Axicons are the most commonly used elements for Bessel beam formation. However, there are also many other approaches demonstrated and being used in applications. Computer controlled spatial light modulators (SLM), for example, can generate the required radially linear phase shift and yield Bessel beams with controllable size and focal depth [18–20]. Using an axicon immersed in an index matching liquid also provides some tunability (through adjusting the temperature and changing the liquid) and also reduces some aberration observed for small-base-angle axicons [21]. Using the liquid-immersion scheme, theoretical profiles can be reproduced by experiments with very close match, as illustrated in Fig. 2.

Tunable acoustical gratings are also able to provide similar tunability [22]. Other methods used in literature include circular diffraction gratings [23], Fresnel axicons [24] and computer generated holograms [25].



Figure 2 Experimental intensity profiles of a Bessel beam, generated by passing a Gaussian beam through a liquid-immersion axicon, demonstrated in [21]. Top: 2-Dimensional transverse distribution. Bottom: Radial distribution, and comparison to an ideal Bessel function.

2.1.2. Airy beams

Airy beams constitute one of the most remarkable results of beam shaping, since they yield the intriguing "accelerating light". The fundamental idea was first laid down by Berry and Balazs in 1979, in the context of Quantum Mechanincs [26]. They show that a wave in the form of an Airy function propagates with acceleration even in free space. In other words, the maxima of probability density follows a parabola, rather than a line, as the particle propagates. This behavior does not violate Ehrenfest theorem, since the acceleration happens only for the maxima, and the averaged probabilities still move linearly. In addition, the wavefunction also keeps its integrity during propagation.

The paraxial wave equation in optics has the same differential equation form as the potential-free Schrödinger equation. As a result, the Airy wavefunctions should yield the same properties for optical fields. The practical difficulty is that, typical light sources available in a laboratory do not have Airy function or similar transverse profiles. The breakthrough came with Siviloglu et al. in 2007 [27]. They show that cubic spatial phase applied in one or two transverse coordinates would Fourier transform to Airy functions. By applying the required phase profiles via SLM, they were ablo to observe Airy function profile and confirm ballistic propagation [28].

Airy beams share some properties of Bessel beams. First of all, they are also diffraction-free, i.e. their profile and intensity does not change by propagation. As with Bessel beams, such idealization requires infinite energies, yet finite aperture and finite energy Airy beams also exhibit similar features within a propagation zone (again, much longer as compared to Rayleigh distances of Gaussian beams) [29]. Airy beams also exhibit self-reconstruction property [30]. The acceleration of Airy beams corresponds to a behavior that the focal spots (defined as the principle intensity maxima) moves along a parabola, as the beam propagates. The behavior is physical since the average intensity still keeps linear propagation. Transverse electric field of Gaussian beams with cubic phase can be expressed as

$$E(x) = E_0 \exp\left[-\frac{x^2}{w_0^2} - i\varphi_3 x^3\right]$$
(3)

where φ_3 is the coefficient of the cubic polynomial phase function. Fourier transformation of Eq. 3 yields

$$E(x') \propto Ai\left(\frac{x'}{x_0}\right) \exp\left(a\frac{x'}{x_0}\right)$$
 (4)

where x' is the conjugate variable and Ai denotes the Airy function. The parameters x_0 and a are related to the width of the central maxima and decay rate of the fringes, respectively. The optical Fourier transform required after the application of the cubic phase can be performed either by propagating the beam to the far field, or simply using a lens [27].

As opposed to Bessel beams, Airy beams are not circularly symmetric, hence they can be independently generated in the two transverse cartesian coordinates. Transverse and longitudinal intensity cross-sections; and full transverse profile of an Airy beam (in accordance with finite energy described by Eq. 4 is given in the Fig. 3.



Figure 3 Theoretical intensity profiles of an Airy beam, generated by passing a Gaussian beam through a cubic spatial phase. Top: 2-Dimensional transverse distribution. Center: Transverse cross-section along one axis. Bottom: Longitudinal-transverse distribution during ballistic preparation. In the calculations, light wavelength is taken as 633nm and a = 0.11.

While the computer controlled SLM yields satisfactory beam profiles, these devices also have limitations. The phase responses of active elements of SLMs usually have strong dispersion and the obtainable phase depth depends on the light wavelength. The voltage-phase characteristics should also be calibrated carefully and corrected in the phase calculations. For practical purposes, it would be highly desirable to have a single passive element that would perform Airy beam transformation. It was shown recently by Yalizay et al. that such an element can be conveniently formed by an arrangement of positive and negative cylindrical lenses [31]. Experimental profiles obtained by this Airy lens closely follow the expected Airy profile and ballistic behavior [31] and the focal points move along a parbola (see Fig. 4). A similar approach was also suggested by Papazoglou et al., which uses the aberrations of cylindrical lenses, when used at large angles with respect to the input beams [32]. A specialized nonlinear optical method was also demonstrated for the same purpose [33]. As Airy beams are introduced to the literature quite recently, generation methods are much limited. Nevertheless, as we summarize below, many applications emerged in a short time period; and due to the intriguing properties of the Airy beams, it can be expected that they will find even broader use in the near future.

2.1.3. Caustic beams

Airy beams are accelerating solutions to the paraxial wave equation. By using caustic-based approaches, the acceleration idea can be extended to non-paraxial regimes. Accelerating beams along arbitrary convex curves was first demonstrated by Greenfield et al. [34]. Remarkably, acceleration of light along almost arbitrary order polynomial curves, as well as exponential curves were demonstrated experimentally [34,35]. The phase functions required for such beam formation is explained in terms of optical catastrophe theory, where the curve followed by the intensity maxima forms a caustic [34].



Figure 4 Experimentally measured focal points of an Airy beam, generated by passing a Gaussian beam through an Airy lens, demonstrated in [31].

In all cases of arbitrary convex trajectories, the transverse profiles are in the form of Airy function. However, the non-diffracting behavior holds for much shorter distances (only a few Rayleigh ranges) and the beam strongly diffracts out of this region. The same approach is also used to send light in circles, with arc of angles up to 60-degrees [36].

2.2. Optical vortices

Optical vortices are one of the mostly investigated classes of beams, due to their fundamental properties and applications. The concept was first introduced ny Nye and Berry in 1974, for waves in general [37]. In context of electromagnetic waves, an optical vortex corresponds to a transverse intensity pattern, in which a zero intensity point is surrounded by finite intensities. In practice, such distributions can be obtained through introduction of phase singularities or what is referred to as "phase dislocations". Points on the transverse plane corresponding to the singular phase yield zero intensity, and as a result, a ring-like beam pattern is observed.

The simplest way of turning a regular (e.g. Gaussian) beam into a vortex is passing the beam through a spiral phase delay element. Such an element would introduce an azimuthal-angle-dependent phase given by $\exp(im\varphi)$, where m is an integer called topological charge and φ is the azimuthal angle in the transverse plane. This helical phase would yield an undefined phase at the center and hence these points generate zero intensity. A very interesting direct consequence of the helical phase is that light with helical phase carries orbital angular momentum quantized by $m\hbar$. This angular momentum is verified experimentally through absorption of light by small particles and observing the conservation of angular momentum [38].

There are many alternative ways of optical vortex generation. Certain orders of Laguerre-Gauss beams, which are fundamentally supported by laser cavities possessing azimuthal symmetry, also describe vortices. Two sample Laguerre-Gauss mode intensity and phase profiles are shown in Fig. 5. Holographical methods are also straightforward and relatively easy to implement. Here, a holographic grating pattern with a singularity at the center (or the "fork" pattern) is used to diffract-off an incoming Gaussian beam or plane wave, thereby generating a vortex [39]. In a more recent work, femtosecond-laser nanostructured glass is shown to generate optical vortices [40]. Here, a radially varying half-wave plate is formed and the superposition of two perpendicular polarization states, going through the bifrefringent nanostructured glass forms the vortex.

Lastly, it is also possible to combine the virtues of optical vortices with diffraction-free-beams, thereby generating diffraction-free vortices. For example, a TEM_{10} Laguerre-Gaussian beam with doughnut shape (see Fig.5) can be passed through an axicon and generate a high order Bessel beam with vortex profile [17]. Alternatively, a Bessel beam generated by passing a TEM_{00} Gaussian beam through an axicon can be passed through a spiral phase plate, and generate a similar structure. This scheme also provides an easier-to-understand interpretation of vortex formation, in terms of geometrical optics. The on axis intensity of a Bessel beam is generated by interference of light rays coming from the opposite sides of the axicon, and these rays regularly describe waves in phase. When a spiral phase is introduced, however, the same waves would be exactly out of phase, causing destructive interference on-axis and hence the vortex. An experimental Bessel-vortex beam profile generated by this later mentioned method is shown in Fig. 6.

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Figure 5 Theoretical intensity profiles for TEM_{01} and TEM_{02} Laguerre-Gaussian laser modes and corresponding azimuthal phase profiles for topological charges m = 1 and m = 2.





2.3. Other beam shapes

In the previous sections, we have summarized some particular beam profiles, which exhibit interesting physical properties. It should be clear that in general, there are unlimited possibilities of beam shapes and through simple experimental implementations involving a computer controlled SLM and some polarization optics, light beams can be tailored almost to taste. A more extensive review of beam shaping can be found in a book devoted to this topic [41].

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Figure 7 Intensity profile of a regular triangle beam at three propagation distances.

We close this part of the review on beam shapes by mentioning two other profiles that we deem noteworthy. A special case of accelerating beams is demonstrated as "regular polygon beams". Such beams have intensity maxima on the transverse plane, which can be located at corners of regular polygons. As the beam propagates, these spots separate from each other in a non-linear fashion, hence the acceleration [42]. The generation can be done by phase-only masks. Sample beam profiles for a regular triangle, at three propagation planes is shown in Fig.7.

Lastly, we briefly mention beams generated by intensity or phase grating patterns. Waves going through periodic arrays (with periodicity greater than the wavelength) are diffracted to different angles. In some cases, especially for certain applications, imposing a grating over the phase profile to be used for beam shaping generates many multiples of the same beam shape. These multiple shaped-beams can be projected on the Fourier plane, thereby providing a platform for multiplexing the process under investigation [20]. Another multiplexing method is to use arrangement of multiple phase elements in an array [43].

3. Applications of tailored light in ultrafast optics and photonics

In the first part of this article, we summarized the general framework of beam shaping and provided particular examples. Beam shaping attracts increasing interest from the Optics research community due to fundamental aspects, as well as potential applications. Indeed, as will be shown below, beam shaping can be very appealing as it significantly facilitates many applications and in some cases, it may even enable interactions which were not possible otherwise.

The general idea of beam shaping is used in quite many works in the literature. We restrict our discussion to studies involving ultrafast optics and photonics (i.e. works performed using picosecond or femtosecond laser pulses).



3.1. Plasma generation

Generation of transient plasma channels in transparent materials (in solid, liquid or gas phase) with ultrashort laser pulses is a well-developed method for fundamental studies of light matter interactions [44], as well as for applications such as wake-field acceleration [45] or pulse compression [46,47]. In these studies, the media is ionized by an intense femtosecond laser, through nonlinear absorption mechanisms such as multi-photon or tunnel ionization. If the power in the beam is sufficient, the self-focusing of the laser may collapse the beam on itself, resulting in generation of a relatively long channel, in which light is self-guided with accompanied plasma [44,48]. This effect is called filamentation and globally several research groups are devoted on this study and applications.

Shaped beams provide useful tools to understand some physical behaviors behind filamentation and plasma generations. A particularly important work was carried out by Polynkin et al. [49], who used high power femtosecond laser pulses formed into Airy beams to ionize air. Due to the parabolic propagation of foci of Airy beams, secondary emission from different longitudinal section of the generated plasma can be mapped to different angles.

Air or other gaseous media can also be ionized using Bessel beams. As compared to filamentation initiated by Gaussian beams, Bessel beams provide ability to better probe some interactions. For example, in a Gaussian beam, entire beam profile contributes to the energy reservoir making up the filament and the beam collapses as a whole. In a Bessel beam, a central maxima is surrounded by many concentric rings and these rings provide the energy to the center at further propagation distances. Therefore, a Bessel beam (compared to Gaussian beam of same total power) would yield a more tenuous plasma (due to the distribution of the total energy over the rings), yet the plasma would be less prone to instabilities [50]. Meter-range plasma channels with uniform free-electron densities are demonstrated [21].

The plasma channels generated by filamentation of femtosecond laser pulses is suggested and developed for many applications. Using terawatt-class lasers, the plasma channels can be formed at kilometer-range distances, in controllable manner [51]. This ability calls for application of this method for artificial triggering and guiding of lightning. Although this method is tested in multi-national experimental campaigns, the results are not satisfactory and no actual triggering of lightning has been achieved to date [4]. The main limitation behind this failure is believed to be the very short lifetime (of the order of nanoseconds) of the laser-generated plasma. In order to extend the lifetime of the plasma, one suggested method is to use a secondary pulse, which has comparable or higher energy with the initial femtosecond pulse, but with a much longer duration (some tens of nanoseconds, for example, for a typical Nd:YAG laser). The second laser would not have sufficient peak power to ionize air by itself, yet it can accelerate the free electrons in the plasma and multiply them through impact ionization, thereby extending the overall plasma lifetime [52].

The application of double-pulse plasma heating method also requires a longitudinal matching of the two beams. Since nanosecod pulses cannot generate filaments, in case of Gaussian beams, the diffraction would yield a strong longitudinal mismatch. This problem is solved by forming the long-pulse into Bessel beams with very long focal lines [53]. Proof-of-principle laboratory results indicate significant enhancement of lifetime, over a large longitudinal portion of the filaments.

The plasma-fronts generated through axicon focusing are superluminal due to the well known scissor effect [54,55]. The superluminal plasma-front can emit secondary Terahertz (THz) radiation with improved efficiency [56]. The low refractive index of the plasma channels can be used to form transient waveguides for guiding high-intensity light [57]. Here, Bessel beams provide extended waveguide lengths.

3.2. Laser material processing

One of the most attractive aspects of femtosecond laser pulses is their ability to process material with unprecedented precision and repeatability. These advantages come from the extreme short durations. First, since the pulse duration is orders of magnitude shorter than heat diffusion times (even for materials with highest thermal conductivities), heat diffusion during light absorption is negligible [58]. As a result, only directly irradiated parts of the material are heated and processed (i.e. melted or evaporated). In addition, since the absorption of femtosecond pulses is governed mostly by nonlinear optical processes, the events are highly deterministic and repeatable [59,60]. Finally, by making use of both properties and keeping the fluence on the material close to ablation threshold, sub-diffraction-limit feature sizes can be obtained [61]. Nanometer-precision micromachining with femtosecond lasers is demonstrated by various research groups [61–63].

While the laser nanofabrication method mentioned above works in principle, there are several practical obstacles. First of all, the method requires near-diffraction-limit, hence high numerical aperture (NA) focusing. Tightly focused Gaussian beams diffract very rapidly and therefore, meticulous longitudinal alignment is necessary. The smallest beam size is obtained only at one focal plane, and at every other plane, the beam is larger. Furthermore, extension of the method to smaller sizes could be thought by using shorter wavelengths (which can be easily obtained through harmonic generation of the femtosecond laser source), yet high NA focusing in the short wavelengths (e.g. UV region) is very problematic due to increased optical aberrations and high costs of corrected objectives.





Figure 8 SEM image of a nanostructured pattern generated by femtosecond laser balation, via Bessel beam focusing.

It is shown recently that, using Bessel beams with very steep cone angles can eliminate all these issues. Due to their diffraction-free nature, Bessel beams keep their size unchanged within the Bessel zone (which can be orders of magnitude larger than Rayleigh ranges of similar-size Gaussian beams). This property is already proven to be useful for surface processing, with longitudinal alignment constraints significantly reduced [64]. Generation of these beams at short wavelength brings no additional aberrations or costs, as long as the axicon is made of a transparent material. Using these principles, nanometer-size features are formed on metal thin films [65]. Example scanning electron microscope (SEM) image of nanostructures formed with Bessel beams is shown in Fig. 8.

Long focal depths of Bessel beams also provide advantages for material processing applications, in which high depth is required. Is is known that at flounce levels slightly below ablation threshold, permanent index of refraction modifications can be obtained in glasses [66–68]. The modified regions can serve as optical waveguides. By using Bessel beams, extended waveguides can be written in glasses without the need for sample scanning, thereby avoiding additional scattering losses [69]. An analogous method can also be used at flounces above the ablation threshold, to form long cavities, suitable for microfluidic or similar applications [70,71]. By exploiting the filamentation stability of Bessel beams mentioned above, high aspect ratio channels with uniform diameters are demonstrated [72].

In other works, long foci of Bessel beams are exploited for deep-drilling in semi-transparent and opaque materials [73]. Although the effect of diffraction free nature of Bessel beams in these applications is not obvious (due to absorption of the rings, the diffractionless propagation would cease inside the cavity), it can be related to waveguide-like propagation of light within the machined cavities [74].

Finally, in the far field, the Bessel beams formed by finite size input beams turn into single rings. While this region is out of non-diffraction region and is not of interest for most cases, it can be useful for micromachinin applications, as a quick and clean cutting of circular shapes. Since the laser power is distributed over a large area, high power lasers, such as CO_2 are required [75].

3.3. Nonlinear optics

The virtues of non-diffracting beams is most appealing for nonlinear optical applications. In most nonlinear optical effects, the strength of the interaction is determined by the region over which light intensity is sufficiently high. Therefore, especially diffraction-free beams are worth attention for investigations in this regard. We will summarize results obtained with shaped beams for some particular nonlinear optical interactions.

3.3.1. Second harmonic generation (SHG)

The lowest order and easiest to observe nonlinear optical effect is SHG [76]. The efficiency of SHG is determined by the input intensity and the propagation range. In an early theoretical study, it was suggested that the Bessel beams should have a greater conversion efficiency as compared to Gaussian counterparts [77]. However, this conclusion is disproved both theoretically and experimentally [78]. The main reason behind this observation is that in Bessel beams, the total

power is distributed over many concentric rings. As a result, beam intensity at the center is much less (typically by < 10%) as compared to a Gaussian beam of the same power. The rise of efficiency due to longer focal depths is not sufficient to compensate for lower on-axis intensities [78].

While Bessel beams are not useful for increasing the conversion efficiency of SHG, they can provide other advantages such as tunability. In the momentum space, Bessel beams are made of wave-vectors distributed on a conical surface. This spectrum of propagation directions favor the phase-matching conditions. Bessel beams may provide phase-matched SHG in cases where it is not possible with Gaussian beams [79].

The ring pattern of Bessel beams mentioned above can also be suitable for self-phase-matching in sum-frequency generation [80]. The phase-matching condition can be satisfied by a wide range of refractive index variations, and in addition, it may compensate for phase mismatch induced by Kerr effects. The theoretical explanation of this behavior is based on expressing a ring beam as a superposition of Bessel beams, thus the conical spread of wave-vectors providing the phase-matching tolerance [81].

3.3.2. Self-focusing and related third-order effects

When the intensity of a light beam becomes sufficiently high, the index of refraction of the host medium becomes intensitydependent [76], following the typical relation

$$n(I) = n_0 + n_2 I (5)$$

where n_0 is the index of refraction in the absence of light, and n_2 is the nonlinear index of refraction coefficient, which is related to the third order susceptibility of the medium. Equation 5 has several consequences, generally described as self-action. For example, self-phase modulation (SPM), resulting from the intensity-dependent temporal phase causes modification of the pulse spectrum during propagation. Similarly, self-focusing results from lens-like spatial phase induced by the transverse intensity profile of the light, on itself. Self-focusing and SPM are the main actors behind the filamentation phenomena discussed above.

Due to their long intense propagation, Bessel beams received much attention for investigation of third order effects. The self-focusing of Bessel beams was first investigated through solutions of nonlinear Schroedinger equation [82]. Since the intensity of the Bessel beam is highest at the central main peak and it gradually decreases in the rings, the self-focusing (or self-defocusing in the case of negative n_2) mainly affects the central portions and the beam contraction of the central spot is observed. The compression of the central spot also causes the decrease of the constrast between zeros of the rigns [83]. However, the nonlinear propagation is stable, as asymptotically the solutions reproduce diffraction-free beam pattern. The stability of nonlinear propagation is further investigated in the presence of nonlinear losses [84]. As opposed to whole-beam collapse observed in Gaussian beams, nonlinear Bessel beams consist of low intensity (hence linear optical) rings, continuously providing energy to the nonlinear central spot, yielding stable propagation. Stable propagation is experimentally observed in several works [85–87].

The self-reconstruction of a Bessel beam, which is a linear optical effect, is also observed in nonlinear-optical regime [88]. Conical beams with a large portion of their azimuth blocked are passed through a nonlinear medium, where Bessel profiles are formed. At the output far field, the ring is observed to be completed (i.e. self recontracted). This observation is attributed to Rayleigh-wing scattering.

A particularly interesting outcome of nonlinear propagation of Bessel beams is the far-field formation of a central spot and appearance of an additional outer ring (Fig. 9. These new features are attributed to the self action of Bessel beams resulting from self focusing [89]. The outer ring corresponds to idler wave in parametric amplification of the new central spot by regular Bessel core.

3.3.3. Stimulated Raman Scattering

Extended intense propagation of shaped beams also yields notable results in nonlinear frequency shifts such as Raman conversion. In an early experimental work, stimulated Raman scattering pumped by a Bessel beam is shown to generate Stokes light emitted on a cone, with conversion efficiency as high as 60% [90]. The behavior of anti-Stokes emission with Bessel beam pumping is investigated by Sogomonian et al. [91]. In the anti-Stokes process, two types of phase-matching is distinguished, involving collinear and conical photons. More detailed studies on phase-matching configurations show that in the case of Bessel beams, transverse phase-matching becomes non-negligible [92]. The observed angular spectra of stimulated Raman signals match the transverse phase-matching predictions, including the cases where coupling of second order Stokes and anti-Stokes components takes place [93].



Figure 9 Far field pattern of an intense femtosecond laser Bessel beam, after nonlinear propagation through Xenon. The colorful central spot appears only in nonlinear propagation regime.

3.4. Biological, medical and related applications

Shaped laser beams found broad use in applications related to biology and medicine, especially in imaging configurations [94,95]. Similarly, beam shaping can also be very advantageous for optical trapping and micromanipulation appications [96–99]. In addition to other aspects, the self-reconstruction properties of particular beam shapes are appealing for study of light-matter interactions in non-homogeneous media [100]. In order to stay within the framework of this review, we only summarize applications which involve or require use of ultrashort laser pulses.

The advantages of ultrashort laser pulses in material processing, mentioned in the earlier sections, are mostly valid also for bio-medical applications. In the cases of laser tissue ablation, while the ablation dynamics are typically more complicated due to the involvement of complex chemical processes, what is said about heat localization an minimal collateral damage are still valid. Similarly, the virtues of shaped beams such as diffraction-free Bessel beams can be transferred to ablation of biological tissues [101]. The reduced alignment constraints of Bessel beams are exploited to develop an experimental platform for cellular transfection, based on axicon-tipped fibers [102, 103].

Another frequently used application of ultrashort laser pulses in biomedical applications is nonlinear imaging. Multiphoton absorption and fluorescence imaging methods provide unprecedented levels of optical resolution and three-dimensional imaging capability. Three-dimensional imaging is typically performed by tightly focusing a femtosecond laser pulse inside the tissue and scanning the sample (or the focus) in three dimensions. This time-consuming scanning requirement can be problematic especially in cases where the temporal evolution of the biological sample takes place on comparable time scales. By using the long line foci of Bessel beams, it is possible to simultaneously image (through two-photon absorption) many axial points, while keeping the lateral resolution unchanged [104]. Similar depth of field enhancements are also observed in three-photon fluorescence [87].

4. Conclusions

In this review, we emphasized that controlling the intensity, phase and polarization of light in space provide researchers a very wide range of possibilities for investigations of either behavior of light in a fundamental level, or light matter interactions targeted for applications. Peciluar behavior of light such as acceleration, diffraction-free propagation, self-reconstruction etc. can be observed through beam shaping. Recent advances in light modulation device technology and optical fabrication are expected compliment beam shaping methods. The advantages brought by particular beam shapes brings about appeal from many applications. There is an extensive literature on the use of structured light encompassing many diverse science disciplines. Here, we mainly focused on applications which directly involve the use of ultrashort laser pulses. Structured-beam femtosecond laser pulses are of particular interest in plasma generation, material processing, nonlinear optics and biomedical optics. It should be reemphasized that applications are not limited to those listed in here. Our intention was to draw attention to some of the particularly interesting results. From this summary, it should be evident that there are many potentials for the use of beam shaping both for fundamental studies and for enhancing or enabling certain light-matter interactions.

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