Independent control of aspect ratios in the axial and lateral cross sections of a focal spot for three-dimensional femtosecond laser micromachining

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New Journal of Physics 13 (2011) 083014 (13pp)
Received 22 March 2011
Published 12 August 2011
Online at http://www.njp.org/
doi:10.1088/1367-2630/13/8/083014

Abstract. We theoretically and experimentally show that independent control of aspect ratios of cross-sectional shapes of a focal spot in both axial and lateral directions can be achieved for three-dimensional (3D) femtosecond laser micromachining by the use of a combination of a slit beam shaping technique and a temporal focusing technique. The simultaneous employment of the spatial and temporal beam shaping techniques allows us to achieve isotropic resolution in 3D space even for an objective lens of low numerical aperture. We also present analytical expressions of the peak-intensity distributions near the focus for the spatiotemporally focused femtosecond laser beams with and without utilizing the slit beam shaping technique.

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In recent years, three-dimensional (3D) micromachining based on femtosecond laser direct writing has attracted significant attention and promises a wide range of intriguing applications in many research fields, for example, microfluidics, micro-optics, micro-electronics, photonics and optofluidics [1–13]. This technique offers a unique solution for true 3D microstructuring in transparent materials such as glass and polymer and enables multifunctional integration in a single substrate. Unlike the traditional planar lithography in which the feature size is only determined by the lateral resolution, spatial resolutions in all the three dimensions in space have to be taken into account in femtosecond laser direct writing to fabricate 3D microstructures. Unfortunately, the focal spot produced by an objective lens naturally has an asymmetrical shape elongated in the direction of propagation, resulting in unbalanced lateral and axial resolutions. To date, several beam shaping techniques have been developed to solve this problem [14–20]. For instance, in the widely used transverse writing scheme (i.e. the writing direction is perpendicular to the direction of laser propagation), by shaping the input femtosecond laser beams with either a pair of cylindrical lenses [14] or a narrow slit [15] placed before the objective lens, one can obtain the circular cross section of a focal spot in the plane perpendicular to the transverse writing direction, whereas the cross sections in the planes perpendicular to the writing direction are still elliptical. Moreover, isotropic 3D spatial resolution in femtosecond laser micromachining has been demonstrated by means of the crossed-beam irradiation method [17], whereas the stringent requirement on the alignment of two objective lenses introduces additional complexity and cost as compared to the beam shaping techniques requiring only one objective lens. Thus, beam shaping techniques that could enable 3D isotropic resolution are still in demand.

Very recently, it was reported that nearly isotropic spatial resolution in 3D femtosecond laser micromachining can be obtained with spatiotemporally focused femtosecond laser pulses [21, 22]. The temporal focusing technique was originally developed by the bio-imaging community for suppressing the background signal in two-photon fluorescence microscopy [23, 24]. In our previous work [21], an objective lens of a relatively high numerical aperture (NA) was used (NA = 0.46), which is important for the production of a nearly spherical intensity distribution in the focal volume. With both experimental evidence and rigorous analytical analysis, however, this article shows that the lateral cross-section (i.e. in the plane perpendicular...
Figure 1. Schematic diagram of side view of the focal system using a combination of the temporal focusing and slit beam shaping methods for femtosecond laser micromachining. G1 and G2 are gratings.

to the propagation direction) of the focal spot will no longer be circular when the temporal focusing technique is employed with an objective lens of a low NA value. Further, we show that a truly isotropic 3D resolution for both high and low NA values can be obtained by a combination of the slit beam shaping and temporal focusing methods. The new technique also allows us to independently tune the aspect ratios of intensity distributions in both lateral and axial cross sections. Here, the axial cross section is in the plane parallel to the propagation direction but perpendicular to the transverse writing direction.

2. Theoretical analysis

2.1. Derivation of the analytical expression of the intensity distribution near the focus in three-dimensional space

A detailed description of the temporal focusing scheme with a circular incident Gaussian beam can be found in our previous work [21]. However, in the present paper, we assume that the incident femtosecond laser pulse has either a circular or an elliptical Gaussian profile with tunable ellipticity; namely, its beam waists along the major axis ($W_x$) and the minor axis ($W_y$) can be either the same or different. As we will see below, if a low-NA objective lens is used in the temporal focusing scheme, 3D isotropic resolution is achievable only when an elliptical input Gaussian beam is chosen. Experimentally, the ellipticity of the Gaussian beam can be tuned with a telescope built by a pair of cylindrical lenses [14] or, more simply, a narrow slit [15, 16], as illustrated in figure 1.

Firstly, assuming that the temporal chirp of the incident pulse is pre-compensated, the normalized light field of a spatially dispersed pulse $A_1$ at the entrance aperture of objective lens can thus be expressed as

$$A_1(x, y, \omega) = \frac{A_0}{\sqrt{\pi \Omega}} \exp\left(-\frac{(\omega - \omega_0)^2}{\Omega^2}\right) \times \exp\left(-\frac{[x - \Delta x(\omega)]^2}{2W_x^2}\right) \times \exp\left(-\frac{y^2}{2W_y^2}\right),$$

(1)

where $A_0$ is the field amplitude, $\omega_0$ the carrier frequency, $\sqrt{2\Omega}$ the bandwidth of the femtosecond pulse measured at $1/e^2$, and $\sqrt{2W_x}$ and $\sqrt{2W_y}$ the beam waists along the major and minor axes measured at $1/e^2$, respectively. Neglecting the high-order chirp induced by the
grating pair, the linear shift of each spectral component at the entrance aperture can be written as \( \Delta x(\omega) \approx \alpha(\omega - \omega_0) \). The details of the derivation of the analytical expression of \( \alpha \) are given in the appendix.

After passing through the objective lens, the light field under the slow-varying envelope approximation can be written as

\[
A_2(x, y, \omega) = A_1(x, y, \omega) \exp\left(-ik\frac{x^2 + y^2}{2f}\right),
\]

where \( k \) is the wave vector and \( f \) the focal length of the objective. Then, after propagating a distance \( z \) from the lens, the light field can be described using the Fresnel diffraction equation, which is

\[
A_3(x, y, z, \omega) = \frac{\exp(ikz)}{i\lambda z} \iint_{-\infty}^{\infty} A_2(\xi, \eta, \omega) \exp\left[i\frac{(x - \xi)^2 + (y - \eta)^2}{2z}\right] d\xi d\eta
\]

\[
= \frac{A_0}{\sqrt{\pi \Omega}} \exp(ikz) \exp\left[-\frac{(\omega - \omega_0)^2}{\Omega^2}\right] \frac{1}{\sqrt{1 - \frac{z}{kz_f} + i\frac{z}{kw^2}}} \frac{1}{\sqrt{1 - \frac{z}{kz_f} + i\frac{z}{kw^2}}} \ldots
\]

\[
\exp\left[-\left(\frac{x^2}{Q_x} + \frac{y^2}{Q_y}\right)\right] \exp\left[-i\frac{(z - f)\Delta x(\omega)^2 + 2fz\Delta x(\omega)}{2P}\right],
\]

where

\[
P = \frac{zf}{k} + i(z - f)W^2_x,
\]

\[
Q_x = \left(\frac{zf}{kW_x^2} + i\frac{z - f}{k}\right) \left(1 - \frac{if}{kW_x^2}\right),
\]

\[
Q_y = \left(\frac{zf}{kW_y^2} + i\frac{z - f}{k}\right) \left(1 - \frac{if}{kW_y^2}\right).
\]

In the above analysis, the zero of the Z-axis is set right behind the objective lens. By performing an inversed Fourier transformation of \( A_3 \), the light field in the time domain can be written as

\[
A_4(x, y, z, t) = \int_{-\infty}^{\infty} A_3(x, y, z, \omega) \exp(-i\omega t) d\omega
\]

\[
= A_0 R(z) \exp\left(-\frac{x^2}{Q_x} \frac{y^2}{Q_y}\right) \exp\left[-\frac{\Omega^2}{4(1 + \psi)(t + f\alpha x/P)^2}\right] \exp[i(kz + \omega_0 t)],
\]

where

\[
R(z) = \frac{1}{\sqrt{1 - \frac{z}{kz_f} + i\frac{z}{kw^2}}} \frac{1}{\sqrt{1 - \frac{z}{kz_f} + i\frac{z}{kw^2}}} \frac{1}{\sqrt{1 + \psi}},
\]

\[
\psi = \frac{1}{2P} \frac{(z - f)\alpha^2 \Omega^2}{2P}.
\]

The peak intensity can then be expressed as

\[
I(x, y, z, t) = |A_4|^2 = \left| A_0 R(z) \exp\left(-\frac{x^2}{Q_x} \frac{y^2}{Q_y}\right) \exp\left[-\frac{\Omega^2}{4(1 + \psi)(t + f\alpha x/P)^2}\right]\right|^2.
\]
The lateral dimensions of the focal spot at the geometric focus can be calculated by
substituting $z = f$ and $t = 0$ into equation (5), and we obtain

$$
W'_x = \frac{\sqrt{2} f}{kW_x^2 + \alpha^2 \Omega^2},
$$

$$
W'_y = \frac{\sqrt{2} f}{kW_y}.
$$

(6)

It is obvious that the size of the focal spot decreases along the $X$-direction due to the term of spatial chirp. The most important information given by equation (6) is that with the temporal focusing, the cross section of the lateral focal plane (i.e. the $XY$-plane) is non-circular if the incident Gaussian beam has a circular beam profile (i.e. $W_x = W_y$). Our numerical simulation implemented below shows that the asymmetry becomes more severe for low NA values. In the meantime, equation (6) suggests that the resolutions in the $X$- and $Y$-directions can be balanced by using a non-circular incident Gaussian beam (i.e. $W_x < W_y$).

Moreover, from equation (5), one can also derive the expression of the pulse width, which depends only on the propagation distance $z$ and reaches its shortest value at the position $z = f$. A detailed discussion on the pulse duration can be found in [23]. However, in the focal plane ($z = f$), there is a time shift $t_0 = \alpha k x / f$ along the $X$-direction, which indicates that the shortest pulses arrive at the focal plane at different times in the $X$-direction but arrive simultaneously in the $Y$-direction.

2.2. Numerical simulations

In this section, we show by numerical simulations that a 3D isotropic resolution can be achieved by combining the slit beam shaping and temporal focusing methods. For the sake of simplicity, we use a slit to control the aspect ratio of the cross section of the incident beam, as shown in figure 1. After passing through the slit, the circular Gaussian can be approximately considered as an elliptical Gaussian beam [15, 16]. Detailed experimental configurations are depicted in [20, 21]. It is noteworthy that the grating pair G1–G2 is ruled with 1500 lines mm$^{-1}$ and the distance between them is set at $\sim 250$ mm, in contrast to our previous work [21].

Firstly, we consider a Gaussian beam with circular cross section. The beam size is chosen to be $W_x = W_y = 1$ mm, corresponding to a lower NA of $\sim 0.1$. For this normal focusing geometry without using either the temporal focusing or the slit beam shaping, the distribution of peak intensity of femtosecond pulses near the focus can be calculated using equation (5) by assuming that $\Delta x(\omega) = 0$. It is not surprising that the peak-intensity distribution at the axial cross section is highly elliptical in this case, as shown in figure 2(a). Note that the axial and lateral scales in figure 2(a) are 10 times those in figures 2(b) and (c) owing to the low NA value. As we can expect, with temporal focusing, the focal depth can be greatly reduced, while the peak-intensity distributions on the $XZ$- and $YZ$-planes are very different, as evidenced by figures 2(b) and (c). Thus, the lateral cross section of the focus will be highly elliptical. This result indicates that the temporal focusing method does not allow us to create a fully 3D symmetrical focus for low-NA objective lenses if the incident Gaussian beam has a circular beam profile.

For femtosecond laser micromachining deep inside the transparent samples, objective lenses of low NA values are required because of their long working distances. In this case, the asymmetric peak intensity distribution illustrated in figure 2 must be corrected for achieving 3D
Figure 2. Numerically calculated laser intensity distributions at the focus produced by an objective lens (a) without and (b, c) with the temporal focusing technique in the XZ- and YZ-planes, respectively. The incident femtosecond laser beam has a size of $W_x = W_y = 1$ mm. Note that the scale in (a) is different.

Figure 3. Numerically calculated laser focal intensity distributions produced by an elliptical Gaussian beam with $W_x = 1$ mm, $W_y = 6$ mm in (a) the XY-, (b) XZ- and (c) YZ-planes, respectively.

isotropic resolution. This problem could be solved with the assistance of the slit beam shaping. As shown in figure 3, when we choose $W_x = 1$ mm and $W_y = 6$ mm, both the axial and lateral aspect ratios of the cross sections of the focal spot can be tuned to be $\sim 1$, thus warranting a 3D isotropic resolution. It should be noted that the beam waists in our simulation are chosen according to our experimental conditions. Furthermore, to achieve a 3D isotropic resolution, one has to carefully choose proper values for both the size and ellipticity of the incident beam. The reason why the axial and lateral aspect ratios of focal spots can vary for different sets of waist sizes that fulfill the ratio $1:6$ is that when the absolute values of the incident beam size change, the NA of the focal system will be changed as well, thus influencing the aspect ratios of the focal spot.

It should be noted that in our previous work [21], we have successfully demonstrated the fabrication of microfluidic channels with circular cross sections oriented in both the $X$- and $Y$-directions with only the temporal focusing technique. This is due to the higher NA value (NA = 0.46) of the objective lens used in that experiment. For comparison, we set the beam size to be $W_x = W_y = 6$ mm (corresponding to a high NA of $\sim 0.6$) in our simulation. The numerical results are shown in figure 4, which are different from the results obtained with
Numerically calculated laser intensity distributions at the focus produced by an objective lens (a) without and (b, c) with the temporal focusing technique in the $XZ$- and $YZ$-planes, respectively. The incident femtosecond laser beam has a size of $W_x = W_y = 6$ mm.

$W_x = W_y = 1$ mm (corresponding to a low NA of $\sim 0.1$) as shown in figure 2, but in good agreement with our previous work [21]. In this situation, the focal intensity profile can be approximately considered as a spherical distribution. Therefore, we conclude that creating a nearly spherical focal volume with the temporal focusing technique alone is only useful for high NA values.

On the other hand, the combination of temporal focusing and slit beam shaping allows us to achieve independent control of the aspect ratios of both axial and lateral cross sections. It can be seen from equations (5) and (6) that the aspect ratio of the axial cross section of the focal spot is mainly dependent on the spatial chirp, which can be controlled by adjusting the distance between the gratings pair. On the other hand, the aspect ratio of the lateral cross section of the focal point can be flexibly controlled by changing the width of the slit. To demonstrate the effectiveness of this method, we calculate the intensity distribution in both the $XZ$- and $YZ$-planes of the focal volume using a fixed value of $W_x$ of 1 mm but various values of $W_y$ of 1, 2, 4 and 6 mm. It is evident from figure 5 that changing the slit width strongly influences the intensity profile in the $YZ$-plane. Therefore, introducing the slit beam shaping method can effectively compensate for the lateral asymmetry of the focal spot caused by spatiotemporal focusing in the case where low-NA objective lenses are used.

3. Experimental

The glass material used in this experiment is a well-known photosensitive glass, Foturan, which is composed of a lithium aluminosilicate doped with trace amounts of silver. During the exposure to the femtosecond laser, free electrons can be generated, which in turn reduce the silver ions to silver atoms. By a successive heat treatment, silver atoms diffuse to form nanoparticles. Due to the plasmon resonance scattering of the silver nanoparticles, the laser modified area appears brown. For information on the other characteristics of this glass, see [5].

With this glass, the 3D intensity distribution of femtosecond pulses at the focus can be recorded and inspected under the microscope (in this case, Foturan glass can be regarded as a 3D photographic recording medium). The femtosecond laser amplifier (Coherent) used in this experiment emits 800 nm, 40 fs pulses with a maximum pulse energy of $\sim 2.5$ mJ at a repetition rate of 1 kHz.
Figure 5. Numerically calculated peak intensity distributions produced by Gaussian beams with different ellipticities, showing the capability of independent control of the lateral aspect ratio. Upper row: the $XZ$-plane; lower row: the $YZ$-plane. In the calculation, $W_x$ is fixed at 1 mm for all the figures, whereas various values of $W_y$ are used: (a, e) 1 mm, (b, f) 2 mm, (c, g) 4 mm and (d, h) 6 mm.

rate of 1 kHz. The energy of the beam is varied by a neutral density filter. A $\times 10$ objective with $NA = 0.25$ is employed. The glass samples can be arbitrarily translated three dimensionally by a PC-controlled $XYZ$ stage with a resolution of 1 $\mu$m.

For comparison, first we inscribe parallel lines in both the $X$- and $Y$-directions in Foturan glass samples by spatiotemporally focusing a circular Gaussian beam. The circular incident Gaussian beam is produced using an annular aperture [21]. The diameter of the aperture is set at 2 mm, corresponding to an NA of $\sim 0.06$. The writing speed is fixed at $100 \mu$m s$^{-1}$ and the average laser power varies from 2.5 to 5 mW. Under the above irradiation conditions, no visible modifications of the glass could be observed under the optical microscope. Then the coupon is subjected to a programmed heat treatment. The temperature is first ramped from room temperature to 520 °C at 5 °C min$^{-1}$ and held at this temperature for 2 h. After the samples are naturally cooled down to room temperature, we polish them and observe the inner areas modified by femtosecond laser irradiation under the optical microscope. As shown in figure 6, the cross-sectional profiles in the $YZ$-plane are much more symmetrical than those in the $XZ$-plane, which is in agreement with the calculated results as shown in figure 2.

It is noteworthy that the calculated spot size in figure 2 is significantly smaller than that of the experimentally observed patterns in figure 6. Several reasons could be attributed to this discrepancy. Usually, the real sizes of the laser-affected zones in the transparent materials are determined not only by the focal spot size, but also by many other parameters such as the pulse energy, scanning velocity, repetition rate and so on. In fact, during the interaction between femtosecond laser pulses and transparent materials such as glass, the free electrons created by
Figure 6. Optical micrographs of the transverse profiles of patterns irradiated with a circular Gaussian beam in the (a–e) $XZ$- and (f–j) $YZ$-planes. The values of laser power are (a, f) 5 mW, (b, g) 4 mW, (c, h) 3.5 mW, (d, i) 3 mW and (e, j) 2.5 mW.

Figure 7. Optical micrographs of the transverse profiles of patterns irradiated with an elliptical Gaussian beam in the (a–c) $XZ$- and (d–f) $YZ$-planes. The values of laser power are (a, d) 4 mW, (b, e) 3.5 mW and (c, f) 3 mW.

Photoionization will diffuse, leading to expansion of the laser-affected zones. In addition, the thermal effect will also cause a modification of the area surrounding the focal spot. It is evident from figure 6 that for the specific focal lens used in this experiment, the size of the modified areas varies in proportion to the pulse energy. Nevertheless, although the sizes are different, the shapes of the experimentally observed spots closely resemble those of the calculated focal spots because the heat transfer in glass is a nearly spatially homogeneous process.

Next, we use a narrow slit to replace the circular aperture. The width and length of the slit are set at 2 and 12 mm, respectively. The patterns are recorded 300 $\mu$m beneath the surface. The average laser power measured after the slit but before the objective lens varies from $\sim 3$ to $\sim 4$ mW. All the other processing parameters remain unchanged. After the same processing procedures as those mentioned above, we investigate the transverse patterns in both the $XZ$- and $YZ$-planes again, as shown in figure 7. It can be observed clearly that the cross-sectional profiles in the $XZ$-plane also become symmetrical due to the slit beam shaping effect, which is in good agreement with the calculated results presented in figure 3.
4. Conclusions

In conclusion, we have shown that the spherical distribution of peak intensity near the focus of femtosecond laser pulses can be achieved by combining the temporal focusing and slit beam shaping. We also present a rigorous analytical expression of the intensity distribution near the focus, which not only significantly shortens the simulation time but also enhances our understanding of the focusing properties of the temporal focusing technique. This technique is particularly effective for the use of objective lenses of low NA values, which are widely used in fabricating structures buried deeply in transparent materials because of their relatively large working distances.

It is noteworthy that by using an objective lens of higher NA value, a higher spatial resolution can be obtained. However, it should be pointed out that it is impossible to achieve isotropic resolution with a single objective lens no matter how high its NA is [25]. Theoretically, to achieve an isotropic resolution, the focusing system must be able to collect all the beams with propagation directions fully occupying a $4\pi$ solid angle, which is beyond the capability of a single objective lens. For this reason, even using an objective lens with a high NA of 1.4 (i.e. almost the highest NA that can be offered by commercially available objective lenses), the achievable lateral resolution is $\sim 200$ nm and the axial resolution $\sim 500$ nm for an illuminating wavelength of $\sim 400$ nm. Therefore, the focal spot is still strongly elongated in the axial direction. Moreover, from the application point of view, the use of high-NA-objective lenses in micromachining is severely limited by the short working distance.

We would like to point out that the simplest application of temporal focusing is the control of the aspect ratio of the focal spot. In such case, the incident pulse is linearly chirped in space before entering the objective lens. In the general sense, the focal spot of femtosecond pulses can be tailored to have a more complex structure if incident pulses nonlinearly chirped in space can be used. The combination of spatial shaping techniques (e.g. the slit beam shaping as shown in this work) and the temporal focusing technique therefore opens up a broad range of possibilities for spatiotemporal manipulation of femtosecond laser pulses in a tiny focal volume with spatial resolutions at the wavelength level and temporal resolutions approaching the pulse duration.

Acknowledgments

The authors thank Dr Han Xu and Dr Hui Xiong from Shanghai Institute of Optics and Fine Mechanics for their assistance with aligning the laser system. This work was supported by the National Basic Research Program of China (grant number 2011CB808100), National Natural Science Foundation of China (grant numbers 10974213 and 60825406) and the Opening Foundation of the Key Laboratory of Infrared Imaging Material and Detectors (grant number IIMD-KFJJ-10-01).

Appendix

This appendix is used to derive the analytical expression of the coefficient $\alpha$ in equation (1). The configuration of the gratings pair is shown in figure A.1, where $i$ is the incident angle, $\gamma$ the first-order diffractive angle, $d$ the distance between the gratings and $\sigma$ the groove density of the gratings. The first-order diffraction must satisfy the following equation:

$$\sin \gamma = \sin i - \lambda / \sigma.$$  \hspace{1cm} (A.1)
The left- and right-hand sides of equation (A.1) can be written, using Taylor expansion to first order, as

$$\sin \gamma = \sin \gamma_0 + \Delta \gamma \cos \gamma_0 + \cdots,$$

(A.2)

$$\sin i - \lambda/\sigma = \sin i - \frac{\lambda_0 + \Delta \lambda}{\sigma} + \cdots = \sin i - \frac{\lambda_0}{\sigma} + \frac{\lambda_0}{\sigma \omega_0} \Delta \omega + \cdots,$$

(A.3)

where $\lambda_0$ and $\omega_0$ are the central wavelength and the central frequency, respectively. It should be noted that in equation (A.2), the approximation $\Delta \lambda \approx -\lambda \Delta \omega/\omega$ is used. Combining equations (A.2) and (A.3), it can be derived that

$$\Delta \gamma \approx \frac{\lambda_0}{\sigma \omega_0 \cos \gamma_0} \Delta \omega.$$

(A.4)

The position of different spectral components along the $X$-direction is given as

$$x = d \sin(i + \gamma)/\cos \gamma.$$

(A.5)

Both sides of equation (A.5) can be written, using Taylor expansion, as

$$x = x_0 + \Delta x + \cdots,$$

(A.6)

$$d \sin(i + \gamma)/\cos \gamma = d(\sin i + \cos i \tan \gamma) = d(\sin i + \cos i \tan \gamma_0) - \frac{d \cos i}{\cos^2 \gamma_0} \Delta \gamma + \cdots.$$

(A.7)
Substituting equation (A.4) into (A.7), we obtain

$$\Delta x = -\frac{d\lambda_0 \cos i}{\sigma \omega_0 \cos^3 \gamma_0} \Delta \omega + o(\Delta \omega) + \cdots.$$  \hspace{1cm} (A.8)

Hence, the coefficient \(\alpha = -\frac{d\lambda_0 \cos i}{\sigma \omega_0 \cos^3 \gamma}\).

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