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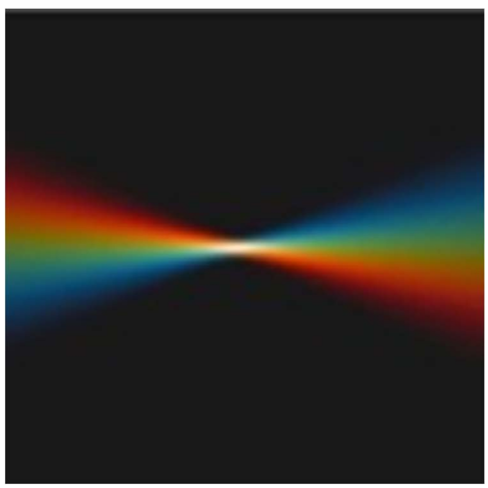

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Breakthroughs in Photonics 2014: Spatiotemporal Focusing: Advances and Applications

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(Invited Paper)

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Abstract: Exploiting the characteristics of spatially chirped ultrafast pulses has been an increasingly active area of research. In this paper, we review the developments in the past year of the microscopy, materials processing, and micromachining fields, where the spatiotemporal structure of these beams is important. We also summarize progress in theoretical and experimental work to better control and characterize spatially chirped beams.

Index Terms: Ultrafast lasers, ultrashort pulse measurements, pulse compression, ultrafast technology, ultrafast optics, nonlinear microscopy.

1. Introduction

As the technology of ultrafast lasers matures, control over the spatio-temporal characteristics of the broadband beams has moved beyond ensuring overlap of the spectral components of the beam to intentionally introducing spatial chirp. This has provided new degrees of freedom to control how these pulses interact with the target. Here, we review the recent advances in the application of angular spatial chirp, which leads to temporal focusing since the second-order spectral phase evolves along the propagation direction [1]. This makes possible depth sectioning in wide-field nonlinear microscopy. Simultaneous spatial and temporal focusing (SSTF) [2] occurs when the frequency components are also focused at the crossing plane. The increased intensity localization has advantages for micromachining and materials processing. The pulse structure is dependent on the system alignment, so techniques for controlling and characterizing these pulses are important. *Transverse* spatial chirp can also be useful: the time-dependent angular rotation of the optical wavefront (the Lighthouse Effect [3], [4]) has been applied to spatially separate attosecond pulses generated in the high-order harmonic process, as described in a recent review [5].

2. Nonlinear Imaging With Temporally Focused Pulses

One of the challenges in nonlinear (two photon excitation fluorescence, second and third harmonic generation, etc.) biological imaging is spanning multiple spatial and temporal scales. A

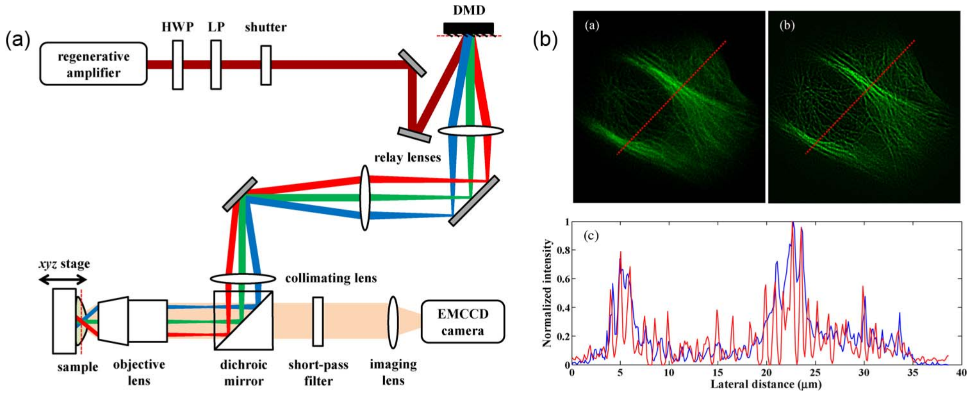

Fig. 1. (a) Temporal focusing setup employing a digital micromirror device (DMD). (b) Comparison of image with temporal focusing alone with an image adding nonlinear structured imaging. Lineout shows the increased resolution [12].

large-field-view is desirable (100 μm to millimeters), with micrometer to sub-micrometer lateral spatial resolution, and micrometer axial resolution. Spatial requirements for an imaging system can therefore span several decades. Dynamic information with sub-millisecond resolution is desirable, along with the ability to track the entire system for extended periods: seconds, minutes or even hours. Thus, temporal requirements for the imaging system also spans decades. A key technology in helping address this range of spatial and temporal characteristics in nonlinear imaging is simultaneous spatial and temporal focusing. As noted above, early work by Oron *et al.* [1] showed that temporal focusing yields a large field-of-view (using low numerical aperture beams or a line cursor as the excitation source) without compromising the axial resolution of the system. The application of temporal focusing to imaging is now one of the most mature aspects of this technology. There were several recent examples in deep (> 500 μm) tissue imaging in *in vivo* samples [6], neuronal networks [7], and biopsy of tissue-cleared samples [8].

The increased dwell time that accompanies these extended excitation source geometries can result in improved signal-to-noise, and consequently allowing faster frame rate and higher dynamic resolution of the nonlinear microscope. Lien and coworkers [9] took advantage of the low trapping force of the temporally-focused beam combined with astigmatism to obtain 14 nm (transverse) and 21 nm (axial) particle tracking resolution at 100 Hz frame rate. An interesting extension of some of the concepts involved in temporal focusing is shown in recent work by Iwai [10]. Using a continuous-wave beam incident on a field with moving objects, they found that the Doppler shift was encoded into the angular spectrum, leading to a technique they call temporal imaging as opposed to temporal focusing.

Improved spatial resolution of SSTF multiphoton imaging was achieved by combining the technique with structured light imaging. For example, a collaboration in Taiwan has demonstrated the use of a digital micromirror device (DMD) to spatially chirp the beam in their multiphoton imaging system in place of the more traditional grating [9], [11], [12] [see Fig. 1(a)]. The programmability of the DMD enabled them to also incorporate structure into the light resulting in measurable gains in the lateral and axial resolution of the system [see Fig. 1(b)]. The same group also implemented an adaptive-optical system with a deformable mirror to optimize the image quality [13].

3. High-Intensity Interactions With SSTF

While the characteristics and advantages of using femtosecond pulses for certain micromachining applications is well known, until the advent of SSTF it was difficult to efficiently ablate or modify a material with a low numerical aperture (NA) beam. In a conventional focusing geometry, the longer confocal parameter can be lead to collateral damage to nearby sensitive structures. Lower NA beams are also susceptible to nonlinear effects such as self-focusing which

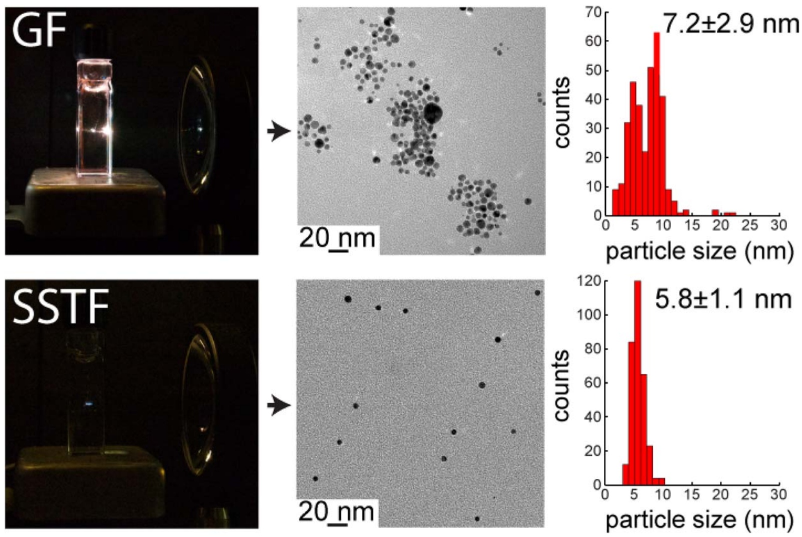

Fig. 2. Comparison of conventional Gaussian focusing (GF) and SSTF focusing into a $10 \times 10 \times 40\ mm^3$ cuvette for producing Au nanoparticles from $[AuCl_4]^-$ in water. Photographs taken at the exposure show strong white light generation for GF and a well-localized spot for SSTF. The resulting particles were imaged with TEM (second column), showing a size distribution (third column) that was much narrower for SSTF [23].

can completely inhibit their application at depth within suitably transparent materials. The strong intensity localization that can be achieved with SSTF allows material modification and interactions either inside the bulk of a sample [14] or on the back surface [15]. By suppressing nonlinear propagation on the way to the target zone, the interaction is much less sensitive to fluctuations in the laser pulse energy. The importance of SSTF for micromachining and materials processing applications were highlighted in three recent review articles [16]–[18].

The capability to modify a material at a specific depth in the material with pulse energy well above the damage threshold is especially important for biological materials [19]. A novel application using temporal focusing was demonstrated recently by a team from MIT and the Harvard Medical School [20]. They used multiphoton excitation for photodynamic therapy to create reactive radical species that induces apoptosis, or programmed cell death, in tumor nodules. By using widefield temporal focusing (700 μm diameter spot) and sufficient energy (55 μJ per pulse at 10 kHz), they were able to perform controlled apoptosis with 10-30 s exposures. By moving to a chromophore with higher two-photon cross-section, they hope to reduce the exposure time to under one second to allow the procedure to be clinically viable.

The localization of the intensity was also demonstrated in a different context (Fig. 2): the synthesis of gold nanoparticles in solution [21]–[23]. By synthesizing nanoparticles in a cuvette, the controlled environment ensures that the composition and purity of the particles is well known. As in the photodynamic therapy application described above, multiphoton absorption leads to the creation of reactive species (H_2O_2) that help reduce $[AuCl_4]^-$ in water after irradiation. With a conventional Gaussian beam focus, nonlinear self-focusing and filamentation clamps the intensity, limits the production of reacting species and produces white-light continuum (Fig. 2), which has been shown to fragment the nanoparticles through surface plasmon resonance heating. With SSTF focusing, the researchers observed a much narrower distribution of particle sizes. The stability and symmetry of the intense focal spot led to rapid radial ejection of cavitation bubbles that effectively mixed the solution.

Along with the strong intensity localization of SSTF, the associated pulse front tilt (PFT) has led to intriguing nonreciprocal writing (or "quill") effects [24], [25]. While more research is needed to better understand these effects, the ponderomotive force of the tilted pulse is an important factor [26]. Recent work [27], [28] contributes to the intrigue. Dai *et al.* [28] observed a coupling of nanograting formation (associated with the beam polarization) with the PFT that depends on the direction of the sample motion. Further evidence of the role of ponderomotive forces in electron transport was observed by Li *et al.* [29], who measured the beam profile of second harmonic light generated by a SSTF pulse in air. The observed symmetric second harmonic profile was consistent with an asymmetric charge separation induced by the tilted pulse. Research with spatially-chirped pulses is also moving into new areas. Zhang *et al.* have proposed a novel quantum optical data storage scheme that takes advantage of the pulse front tilt [30].

4. Generation and Characterization of Spatially-Chirped Broadband Pulses

To take full advantage of the strong intensity localization and pulse front tilt that SSTF makes possible, it is important to understand how the structure is affected by misalignment and distortion, and even more important to be able to characterize the experimental conditions. Several papers investigated further theoretically and computationally the nature and structure of the temporal focus [31]–[33]. Lesham considered the conditions under which the temporal focal plane can be shifted by dispersion for shaped or structured pulses [31]. He *et al.* showed that in addition to the pulse front tilt, the off-axis dependence of the spectral phase leads to a tilt in the plane where the pulse is optimally compressed [32]. This effect can be important for wide-bandwidth and large angular chirp.

Much of the work in materials processing with SSTF has made use of compressors retrofitted into existing systems. In collaboration with KMLabs Inc., our group at CSM developed a Watt-level 10 kHz repetition rate Yb:CaF2 laser system [34]. The chirped pulse amplifier has an integrated single-pass transmission grating compressor, which has improved efficiency compared to previously reported SSTF designs. The efficiency combined with the high-repetition rate of the laser further optimizes its capability for rapid, large volume processing. We also designed a flexible SSTF compressor that allows smooth control of the pulse front tilt [35]. This compressor is a double pass design with unequal paths on each pass, resulting in an output beam with collimated spectral components of variable width. A compact beam delivery system capable of delivering spatially-chirped pulses for endoscopic imaging was demonstrated by Choi *et al.* [36]. They utilized a gradient-index objective to project the spatially chirped beam through a dichroic beamsplitter that allowed the emitted light to be collected with a separate objective.

To fully understand and optimize nonlinear interactions with materials, it is important to align the laser system in a controlled manner and to characterize the spatio-temporal properties of the beam. Much of SSTF work employs off-axis parabolas, but in many applications the use of lens systems is more convenient. Sun *et al.* studied the effect of Zernicke phase aberrations on the quality of the focal intensity distribution [37]: Many of the common aberrations will affect the spatio-temporal quality of the focus, since frequency components travel through different parts of the aperture. In extreme cases, even using an off-axis parabola can lead to unexpected distortions [38]. Our group has developed a spectrally-resolved knife edge scan technique for characterizing the focus in situ and a spectral interferometric technique for characterizing the parallelism and divergence of the frequency components [39]. Wang *et al.* developed a technique for characterizing the pulse front tilt interferometrically by using a non-tilted reference pulse [40].

More general diagnostic techniques, useful for arbitrary beams, were improved upon in 2014. Gallet *et al.* [41] have further refined the SEA-TADPOLE technique [42] in which a single mode fiber is scanned across a plane and interferometrically compared to a reference signal. For spatio-temporally complex beams that might emerge from a nonlinear process, a single-shot technique for producing interferograms at many spectral slices can be used [43], [44].

5. Conclusion

The use of spatially-chirped beams for microscopy, micromachining and nonlinear optics continues to become more widespread. At this time, wide-field nonlinear microscopy is the most mature of the areas of application. Temporal focusing is now being combined with other techniques for improving image resolution such as structured illumination. As the interaction of these beams with materials becomes better understood, simultaneous spatial and temporal focusing is being exploited for its intensity localization and for the material modification effects made possible by the tilted pulses fronts. The ongoing work to develop SSTF delivery systems and characterization methods will help lead to better control and understanding of the processes driven by SSTF pulses.

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