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Vortex Lasers and Sensor Manufacturing (VORTEX-SENSORS)

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ABSTRACT

Development of nanofabrication methods are essential for future-generations of sensing technologies, consumer electronics, and personalised medicine, amongst others. This project brings forwards a new structured laser technology and process know-how to advance nanometre scale (1-100 nm) laser-based manufacturing with feature sizes two or more orders of magnitude smaller than the wavelength of light. We demonstrate the world-highest power Q-switched vortex laser and operating at high pulse rate. This turnkey system with robustness and power-scaling potential can enable industrial scale laser nanofabrication and other high-power structured light applications.

Keywords: Lasers; vortex light; structured light; manufacturing; nanofabrication.

1. INTRODUCTION

Manufacturing at the nanometre scale (1-100 nm) is an essential bridge to translate nanoscience research into practical nano-technology devices and is pivotal for future-generations of sensing technologies, consumer electronics, and personalised medicine, amongst others. Nanofabrication methods exist but lack some flexibility requiring complex and expensive fabrication facilities. Lasers provide flexible, low cost, precision processing tools using material ablation, but with micron scale light wavelength they would appear to have no capacity for nanofabrication. Recent results have shown, somewhat surprisingly, structured laser light can fabricate nanosized features orders of magnitude smaller than the wavelength of light, including tall high aspect ratio nanoneedles with nanometre scale tips, and even having helical external profiles using vortex light (see Fig. 1). Controversy still exists about the precise mechanism(s) for the nanometre scale profile formation and to date processing has been limited to very low pulse rates.

This project makes advance in laser-based manufacturing by addressing critical aspects of laser technology required for nano-manufacturing to be transformed to a commercial or industrial scale: i) a new scalable structured-light laser technology is developed with future potential for operation even at the industrial (kW) power scale; ii) theoretical analysis of the underpinning mechanisms for nanofabrication; and iii) application of the structure light source to perform direct surface nanostructuring trials on thin films at high pulse rates. The output of this project includes the demonstration of the world-highest power Q-switched vortex laser. It is developed with a robust all-solid-state format and has high pulse rate as required for high throughput nanofabrication. The trialling of structured light nanoprocessing demonstrations is underway with fundamental new understanding of structured light nanoprocessing underpinned by theoretical work. We are on a development path of our innovative structured light technology to higher industrial (kilowatt) scale powers and in commercial platforms. The laser technology and process know-how will enable a world-leading position in laser-based nanoscale manufacturing and provide new paradigms for laser nano-printing of 3-D nanostructured devices and new sensory nanophotonic materials with features controllable even with pulse-to-pulse placement flexibility.



Fig. 1. Nanofabrication performed by vortex light carrying orbital angular momentum transports molten material into central (dark) phase singularity where sub-wavelength-sized chiral nanoneedle is created.

2. STATE OF THE ART

Sensor technology impacts almost all major technologies and new and improved sensor systems will continue to be needed in the future based on increasingly small nanometre scale structures. Photolithography, electron beam lithography, and chemical synthesis are used for nanofabrication and whilst they will underpin mainstream nanotechnology, they lack some flexibility and often require complex and expensive fabrication facilities. Alternatively, lasers are flexible, precision material processing tools, playing a key role in industrial manufacturing of a wide range of materials and devices across all industrial sectors. However, material processing by laser ablation operates with optical beam size fundamentally limited by diffraction to the micron wavelength scale of light, and this suggests lasers are ineffective for nanometre scale fabrication. This is a great loss, as laser manufacturing is an extremely versatile and re-configurable tool for material processing and a flexible complement to photolithography.

Recent results show, however, that laser nanostructuring with feature sizes orders of magnitude smaller than the wavelength of light is indeed possible by using *spatially* structuring laser light and delivery in a short pulse duration of appropriate energy. Nakata [1] projected a lattice pattern with a femtosecond laser pulse (λ =785 nm) on a gold film to produce a 2-D array of nanoneedles with tip radius of curvature 3.4 nm. Toyoda [2] further showed vortex beams with nanosecond pulses (λ =1064nm), create tall chiral metal nanoneedles in the vortex dark core with helical external profiling, and tip radius of curvature < 40 nm. No prior known technique can twist metal on a nanoscale. Toyoda's explanation for the twisting of the chiral nanostructure was transfer of orbital angular momentum of the vortex light to the melted metal. However, this explanation has been challenged by Syubaev [3] who showed even higher quality chiral nanoneedle creation and smaller tip radius ~ 10 nm with structured light having no vorticity and proposing a mechanism of temperature-gradient-induced mass transfer. Controversy still exists of the exact mechanism for nanofeature fabrication and whether different mechanisms dominate in different regimes.

Prior structured light nanofabrication has only been performed at low power and low pulse rates (e.g. Toyota et al operated at 10 Hz [2]) unsuitable for commercial or industrial manufacture. To date vortex beams are mostly created by bespoke techniques outside the laser using elements with limited power handling and high expense (e.g. spatial light modulators) or elements with low efficiency or imperfect quality. Direct vortex generation from laser would provide a more commercial turn-key solution but to date Q-switched vortex lasers have only operated to $\sim 1W$ power [4], whereas industrial manufacture will require high powers (even to kW) and high pulse rates for high throughput nanofabrication.

3. BREAKTHROUGH CHARACTER OF THE PROJECT

Controlled manufacturing of features with nanometre size (1-100nm) directly from a laser would be a disruptive due to technological advance flexibility and programmability of the laser (placing nanofeatures even on a pulse by pulse basis) and in relatively low cost, small footprint production systems. Requirements critical for the advancement of laser-based nano-manufacturing include: i) a fit-for-purpose laser technology; ii) a sound understanding of nanoscale light-matter mechanisms to address what nano-features are able to be fabricated; and iii) well-managed protocols ("recipes") for laser nanofabrication tools and process control operating at high throughput. This project makes advancement in many of these key requirements.

We have advance transformative vortex laser and structured light technology in this project as a turn-key fitfor-purpose laser solution. We demonstrate a radically new high-power vortex laser technology that can convert any fundamental mode laser into a vortex laser by replacing its standard output coupler (OC) mirror with what we call a "vortex output coupler" (VOC). Using this technology, our project delivers the world-record highest power vortex Q-switched laser, to best of our knowledge. The laser technology is developed with a robust all-solidstate format and with high pulse rate (multi-hundred kHz) which is important for high throughput nanofabrication. We are already engineering our own higher power versions of this structured light technology and there is no reason to believe we cannot scale to industrial (kilowatt) scale powers and in fully commercial platforms.

We have also created a second structured light innovation where we convert our vortex laser technology into a new spiral "intensity" light structure, that to our knowledge has never been exploited previously as a processing structured light pattern. The spiral intensity structured light can be produced as a simple enhancement to our vortex laser technology.

Our project advances fundamental understanding of the structured laser nanoprocessing by the creation of gradient forces that can operate on sub-wavelength scales. We are set-up to begin laser nanofabrication trials of thin film samples.

Nanostructures created by our vortex and structured light technology would have many potential applications including for sensing. Chiral nanoneedles using metals with plasmonic properties have potential to selectively distinguish chirality and optical activity of molecules and chemical compositions and perform nanoscale imaging in atomic force microscopy and scanning tunnelling microscopy. Vortex light nanoprocessing of monocrystalline silicon has application in silicon photonics, photovoltaics and nano- and microelectromechanical systems, but other possibilities exist. Growing large areas of nanostructured arrays could create new smart sensing materials at high speed and low cost, and as a surface profiling method these might also be flexible or wearable. Structured light nanoprocessing across many classes of materials (e.g. metal, crystal, dielectric, polymer) will create a new fabrication paradigm for next-generation sensors and related devices for various market sectors (e.g. medical, environmental. energy), complementing rather than competing with other existing nanotechnology techniques.

4. PROJECT RESULTS

Our project results have advanced critical aspects needed for structured light nano-feature manufacturing, including structured light technology advancement and process know-how with underpinning fundamental light-matter understanding.

4.1 Structured Light Technology.

We bring forwards both a new vortex laser technology and additionally a new *spiral intensity light* structure in fit-forpurpose turn-key format needed for nanofabrication on a commercial scale. Our new high-power vortex laser technology is based on a patented technique that can convert any fundamental mode laser into a vortex laser by replacing its standard output coupler (OC) mirror with what we call a "vortex output coupler" (VOC). The VOC is a robust common-path Sagnac interferometer with controlled misalignment imbalances converting a Gaussian mode into a high quality (LG₀₁) vortex output carrying orbital angular momentum, (see Fig.1).

Fig. 2 shows schematically our first proof-of-concept implementation of the vortex laser. A "standard" diodepumped Nd:YVO₄ laser operating at 1064nm and fundamental TEM₀₀ mode is Q-switched with an acoustooptic modulator (AOM) as shown in Fig. 2a. This laser was converted to a Q-switched vortex laser as shown in Fig.2b simply by replacement of the output coupler (OC) by our vortex output coupler (VOC). Fig. 2c shows power output (red points) of our proof-of-concept vortex laser operating at > 5W, and pulse repetition rate 150 kHz. Fig. 2c also shows for comparison the power curve (black points) for the standard TEM₀₀ (non-vortex) laser. One impressive achievement is that the vortex laser runs with almost identical efficiency to the standard laser i.e. replacement of output mirror for the vortex output coupler (VOC) is near lossless. The LG₀₁ vortex mode quality was analysed and shown to be near perfect and its handedness was switchable with a single simple control in the VOC. To date other pulsed direct vortex laser technology, to our knowledge, has been limited to low power ~ 1W level [4], so even this first proof-of-concept laser is already worldrecord for direct laser vortex output. Other vortex laser methods try to "force" the laser into a vortex mode that becomes unsustainable as thermally induced lensing and aberration develop at high powers. Our vortex

methodology does not have this limitation, as the internal mode is maintained as a fundamental mode. A further desirable discovery of the VOC was its ability to filter the intracavity mode to maintain TEM_{00} even beyond what the standard cavity could maintain. The details of this work are published in the peer-reviewed literature [5].



Fig. 2. a) standard Q-switched diode-pumped Nd:YVO4 laser; b) conversion to vortex laser with use of vortex output couple (VOC); c) power curve of standard and vortex Q-switched laser; d) power scaled vortex laser to > 14W.

Fig. 2d shows results of further scaling we have performed subsequently taking power to ~14 W using an end-capped Nd:YVO₄ crystal to mitigate some thermal impact on the crystal, the results of which were presented at Photonics West, 2020 [6]. In most recent work, using more efficient in-band diode pumping (880nm rather than standard 808nm diode pumping) we have further scaled power to > 25W. It should be noted that the challenge in power scaling was for generating the TEM₀₀ laser, rather than its conversion to a vortex laser. With operation in high power industrial class laser technology (e.g. thindisc or fibre laser) we see no limit to vortex power scalability, as the elements of the VOC are all high-power handling and with minimal loss. Industrial-class power scaling will enable high-throughput manufacturing of nanostructures with flexibility and low cost.

A second structured light source was also produced by a simple conversion of the vortex laser into a *spiral intensity light pattern* (see Fig. 3). We interfere some of the mutually coherent Gaussian output from the rear laser mirror with the vortex light from the VOC at a 50% beamsplitter. Varying the relative radius of curvature of the beams leads to controlled helicity of the spiral light. To our knowledge, this is the first high power generation and material processing application of a spiral intensity light structure. This spiral intensity will lay down heat driven pattern in the material to be processed to drive strong vorticity gradient surface tension forces that are even stronger than optical orbital angular momentum.



Fig. 3. (left) Vortex intensity pattern from VOC; (right) spiral intensity light pattern produced by interfering vortex output with a Gaussian beam (from intracavity mode).

4.2 Nanofabrication Process Understanding and Know-How

We have developed theory for the structured light processing based on the hypothesis that a focused vortex laser beam with appropriate pulse energy melts (rather than ablates) the material in its high intensity annular (micron scale) region. The melted material is transferred to the dark vortex core by the surface tension gradient $\nabla \sigma(\mathbf{r})$ (Marangoni effect [7]) caused by the spatial temperature profile T(r) produced by the pulsed spatially structured laser heating distribution q(r). A key piece of understanding is that since the surface tension σ of metals (and many other materials) decrease with temperature $(d\sigma/dT < 0)$, hot molten material is pulled by surface tension to cooler regions. For the case of the vortex light, this causes rapid influx of molten material into the dark vortex core which is an order of magnitude smaller than the overall optical mode size and with a steep gradient between the peak intensity and the central zero. The influx of matter can grow tall nanoneedles as towering structures narrowing to nanotip feature size in the range of a few nm. The vortex beam or spiral intensity beam can also impart rotational spin (orbital angular momentum) to the molten matter creating chiral (helical) structuring and provide stabilising rotational symmetry to the needle growth. We have developed theory of the Marangoni surface tension forces alongside the start of numerical simulation of the fluid-dynamics Navier-Stokes equations for molten metal flow to understand and control the underlying nanoscale mechanisms at ultra-high pulse rates needed for future industrial scale manufacturing. The trialling of structured light nano-processing demonstrations is ready to be undertaken on thin-film metal coated substrates using a three-axis beam scanning system with motion control where an additional quarter wave plate combines spin angular momentum that adds (or subtracts) to the orbital angular momentum. It provides some further understanding of the relative mechanism underlying the chiral forces in the vortex processing.

5. FUTURE PROJECT VISION

The future project vision is to develop a major stepchange both in structured light technology and in the laser-based nanofabrication.

5.1. Technology Scaling

Our ground-breaking structured light technology has a current TRL about 3. Our future vision is to develop its power and performance, alongside operation towards commercial platforms. A major vision is to develop a laser roadmapping and path to commercial products to meet current and future market needs. We can in-house develop systems to TRL 5, but will look at collaborations / commercial interaction with laser companies to accelerate TRL status. These laser developments will underpin commercial/industrial applications of structured light.

The second and project specific future project vision is the development of laser nanofabrication. This will demand a concentrated programme of theory (both analytical and numerical simulation) to understand underpinning science of the light-matter gradient processes operating at subwavelength scales. The other aspect to be developed is the experimental nanofabrication tests to develop process know-how. The combined theoretical and experimental work will provide fundamental limits of features sizes; understanding laser energy scaling and material volumetric requirements; nano-feature shapes; best materials to use and substrates to operate upon; preshaped material sub-structures (e.g. islands of materials; tramlines of material) that will control material flowpaths: formation of arrays of material bubbles and other shapes at nanoscale sizes. These developments will ultimately need the input of industrial players in these fields.

5.2. Project Synergies and Outreach

We have a strong laser and photonics core team at Imperial College London and strengthened by the engineering expertise of Unilase Ltd to translate scientific developments into engineered prototypes. We also will need to build a greater critical mass of personnel for our project in Phase 2, to meet the greater technical and market-driven challenges and to address a wider range of new opportunities. We would welcome the input of other collaborators to this project to support its technical success: both other academic groups, and collaboration with other businesses, especially laser companies, and to identify alliances with players in the manufacturing sector (academic-based centres and corporate players). Synergy with other companies in the sectors will provide us with better understanding of market needs for market-driven product development, as well as access to market opportunities. We would look at outreach public dissemination at conference etc but would establish a website presence with product information, news items and white papers.

5.3. Technology application and demonstration cases

In Phase 2, we will provide robustly packaged structured light laser technology demonstrators at increasingly higher powers, pulse rate and TRL level. An advanced laser-based processing system will be established for experimental in-house trials with nanometre precision motion control in 3-D. Nanofabrication trials will initially be conducted in metal thin-films, particularly gold and silver and materials with known plasmonic properties. Dedicated numerical modelling will be conducted to simulate laser-matter interaction at the sub-wavelength scale. This project will meet policy priorities of the Europe 2020 strategy by addressing the challenges to benefit European society and their citizens in medical devices for "Health and wellbeing"; in industrial nanofabrication for "Secure, clean and efficient energy", "Smart, green and integrated transport"; "Climate action, environment, resource efficiency and raw materials".

5.4. Technology commercialization

We are keen to develop the structured-light technology to enable commercialisation. Our consortium team has intellectual property (IP) in the form of patent at international stage. We have gained considerable knowhow and product prototyping development, that form a good starting base to build towards commercial opportunity. The PI and consortium members on this project already has experience of Spin-Out and successfully taking a laser technology from research lab to market. We have proven ability to raise the TRL of the laser technology but forming commercial product is difficult and strategic alliance with an appropriate laser company would accelerate our progress. We have market experience in the "traditional" laser manufacturing sector but lack some experience in the nanotechnology and nonlaser-based manufacturing sectors. We would encourage interaction and alliances in this sector. With an increased team we aim to address a wider structured-light development to address other market opportunities. We propose to look with our in-house Technology Transfer organisation at Imperial at routes to exploitation. This might be via licence, but Spin-Out company is an option as it might better capture the full value of a platform technology applicable across multiple market sectors.

5.5. Envisioned risks

Risks exist by the very nature of new innovative technologies, both at a technical level and at a market level. Key technology risk is not providing suitable competitive performance for the application, which we would mitigate by our increased team to resolve issues. Due to the wide application opportunities if one performance goal cannot be met to focus effort onto another opportunity/application.

Liaison with Student Teams and Socio-Economic Study

As a University research group, we already have wide experience of undergraduate and MSc projects and have indeed incorporated these into the Phase 1 project. We believe this light technology is very accessible for MSc student team projects and encourage projects from basic research, to evaluation of prototypes through to market research in key potential markets and applications. We would appoint a key experienced person on our consortium team to lead the interaction with MSc teams.

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