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Short pulse laser micro/nano manufacturing: fundamentals and applications

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Abstract: Lasers, especially femtosecond lasers, open wide-range and exciting new possibilities in micro/nanomanufacturing of metals, polymers, semiconductors, ultrahard materials, transparent materials, and tissues. This paper reviews the fundamentals, applications and challenges of laser micro/nano manufacturing, including ultrashort laser materials interaction, laser direct-writing manufacturing, laser interference manufacturing, laser near-field manufacturing, laser manufacturing of nanoparticles and nanotubes, and laser assisted diamond crystals growth.

Keywords: laser materials interaction; laser direct-writing manufacturing; laser interference manufacturing; laser near-field manufacturing.

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1 Fundamentals

A femtosecond laser can fully ionise almost any solid material with greatly reduced recast, microcracks and heat-affected zone (Jiang and Tsai, 2003). Compared with a long pulse, a femtosecond pulse in some aspects fundamentally changes laser materials interaction mechanisms. New phenomena induced by femtosecond lasers lead to a new area of ultrashort science. While there is a growing body of experimental observation, a comprehensive model remains undeveloped (Wolff-Rottke et al., 1995; Jiang et al., 2009), of which metals and dielectrics are significantly different with each other (Anisimov et al., 1974; Gamaly et al., 2002; Gan and Chen, 2009; Hertel et al., 1996; Jiang and Tsai, 2005a; Qiu and Tien, 1992; Rethfeld et al., 2002; Kaiser et al., 2000; Petrov and Davis, 2008; Rajeev et al., 2009). It is a significant challenge to explain phenomena associated with complex non-equilibrium and non-linear processes.

1.1 Femtosecond laser metals interaction

The energy transport process of femtosecond laser heating of metals can be theoretically divided into two stages (Qiu and Tien, 1993; Qiu et al., 1994; Tzou et al., 2002):

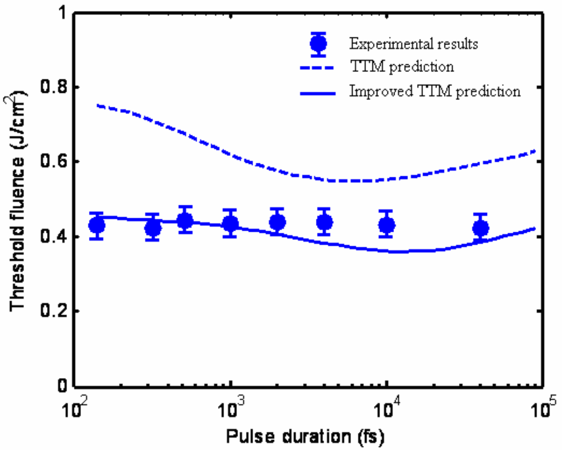
1. laser energy absorption through photon-electron interactions within the pulse irradiation
2. absorbed energy redistribution to lattice through electron-phonon interactions after the pulse duration, for which, the two-temperature equation was proposed and is being widely used:

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \nabla\left(k_e(T_e)\nabla T_e\right) - G(T_e - T_l) + S(z,t) \tag{1}$$

$$C_l(T_l)\frac{\partial T_l}{\partial t} = G(T_e - T_l) \tag{2}$$

Tien's group (Qiu and Tien, 1992, 1993; Qiu et al., 1994) made pioneering theoretical and experimental contributions in this area. However, in their model, the estimations of some key thermal and optical properties are limited to electron temperatures much lower than the Fermi temperature. Jiang and Tsai (2005a, 2007) improved the two-temperature model (TTM) by introducing quantum treatments for key parameter calculations, which made the estimations valid for any electron temperature. Figure 1 shows the damage threshold predicted by the classical TTM and the improved TTM.

Figure 1 Damage threshold fluences of 200 nm gold film processed by a 1,053 nm laser (see online version for colours)



Source: Jiang and Tsai (2005a)

1.2 Femtosecond laser dielectrics interaction

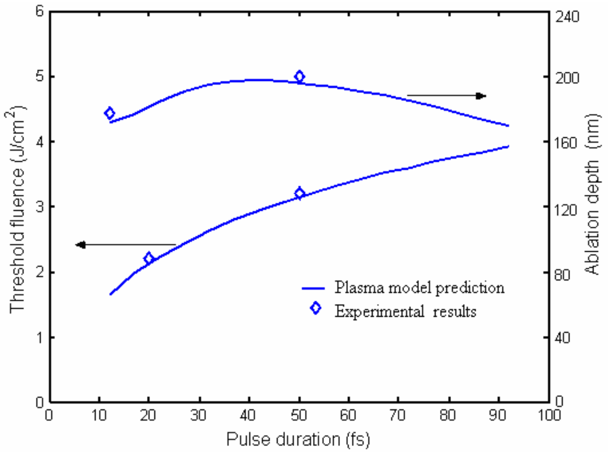
For dielectrics, building up of electrons is crucial to initialise ablation. Once free electron density reaches a critical value, dielectrics are transformed into absorbing plasma with metallic properties, and subsequent laser plasma interaction causes ablation (Kaiser et al., 2000). The energy transport process can be divided into two stages (Ladieu et al., 2002; Petrov and Davis, 2008; Rajeev et al., 2009):

1 Photon energy absorption by electrons, which includes linear process (free electron heating) and non-linear process (bound electron excitation and ionisations) (Du et al., 1994; Perry et al., 1988; Stuart et al., 1995). In an excitation process, valence electrons are excited by photons to higher quantum energy levels within the valence band. In an ionisation process, a valence electron is freed from an atom (or a molecule), where avalanche ionisation (power density ~10^{12} W/cm^2), multiphoton ionisation (~10^{13} ~ 10^{14} W/cm^2), and tunnel ionisation (>10^{15} W/cm^2) are the major competing mechanisms.

2 Absorbed energy redistribution to lattice leading to material removals, which can be divided into thermal process (melting and vaporisation) and non-thermal process (Coulomb explosion and electrostatic ablation) (Nakamura et al., 2009; Stoian et al., 2000; Toulemonde et al., 2000). The competing phase change mechanisms may coexist and/or transit to each other during dielectrics ablation process.

Based on the Fokker-Plank equation and experimental data, a flux-doubling model was developed for free electron generation for the femtosecond laser ablation of dielectrics (Stuart et al., 1995). Jiang and Tsai (2005b) proposed a plasma model with quantum treatments. Figure 2 shows the threshold fluence and ablation depth predicted by the plasma model. Besides, the plasma model also is used to predict some of unexpected phenomena such as flat-bottom crater shapes formed by a Gaussian beam, which were experimentally validated (Heltzel et al., 2007).

Figure 2 The threshold fluence and ablation depth (see online version for colours)



Source: Jiang and Tsai (2005b)

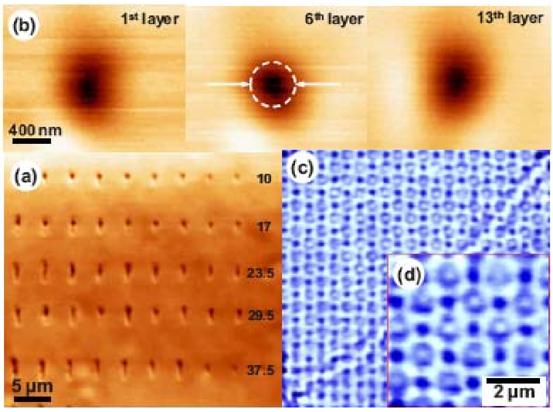
An integrated multiscale physico-chemical modelling shall be established to understand the ultrashort, non-equilibrium and non-linear laser-materials interactions from nanometre to millimetre and from femtosecond to microsecond. Mechanisms of material removals by femtosecond laser remain controversial. The particular mechanism strongly depends on material properties, laser fluence, pulse duration, wavelength, and number of pulses. Incubation and electron screening can significantly reduce the probability of Coulomb explosion. It remains a challenge to clearly explain the transition from one phase change mechanism to another and predict the roles of the competing ablation mechanisms.

2 Processes

2.1 Laser direct-writing micro/nano manufacturing

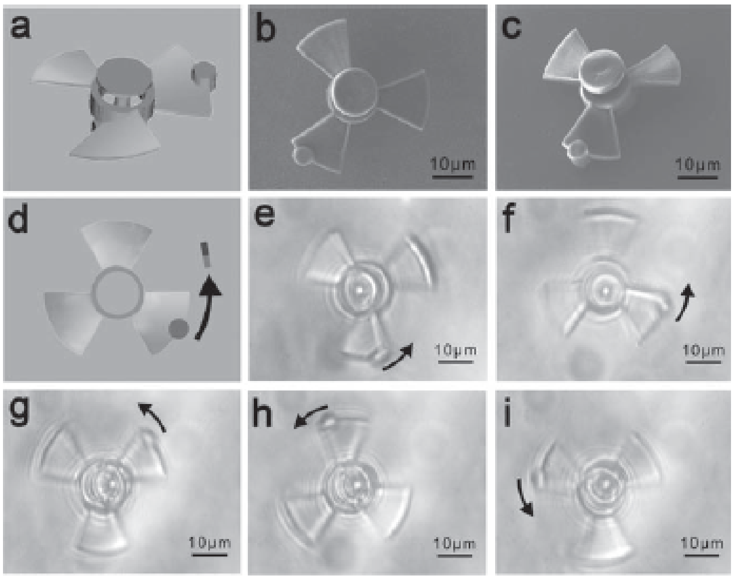
Laser direct-writing is one of the most important manufacturing methods for 3D micro/nanoscale structures and devices (Masuda et al., 2003). Compared with photolithography manufacturing, laser direct-writing is fast, simple and flexible. Especially, by using unique multiphoton absorption and the corresponding strong threshold effects, femtosecond laser processing of dielectrics can achieve sub-diffraction-limit real 3D structures (Joglekar et al., 2003). Various structures and devices were fabricated on or inside glass, polymer and transparent materials, including photonic structures (as shown in Figure 3, Rodenas et al., 2008), nanovoid structures (Kanehira et al., 2005), optical waveguides (Uppal et al., 2008), couplers (Kowalevicz et al., 2005), optical gratings (Beresna and Kazansky, 2010), and optical amplifiers (Psaila et al., 2007).

Figure 3 Laser direct writing of a 3D photonic structure (see online version for colours)



Source: Rodenas et al. (2008)

Figure 4 Laser direct writing of a micro-fan



Source: Xia et al. (2010)

Multiphoton polymerisation can achieve high quality structures and devices with almost any 3D structures (Maruo et al., 1997; Cao et al., 2009). For example, a bull sculpture of 10 µm length and 7 µm height was fabricated with a sub-diffraction-limit spatial resolution of 120 nm (Kawata et al., 2001). Further, micro-machines such as micro-springs and micro-turbines were fabricated and remotely controlled by using photopolymerisable ferrofluid (Xia et al., 2010), as shown in Figure 4.

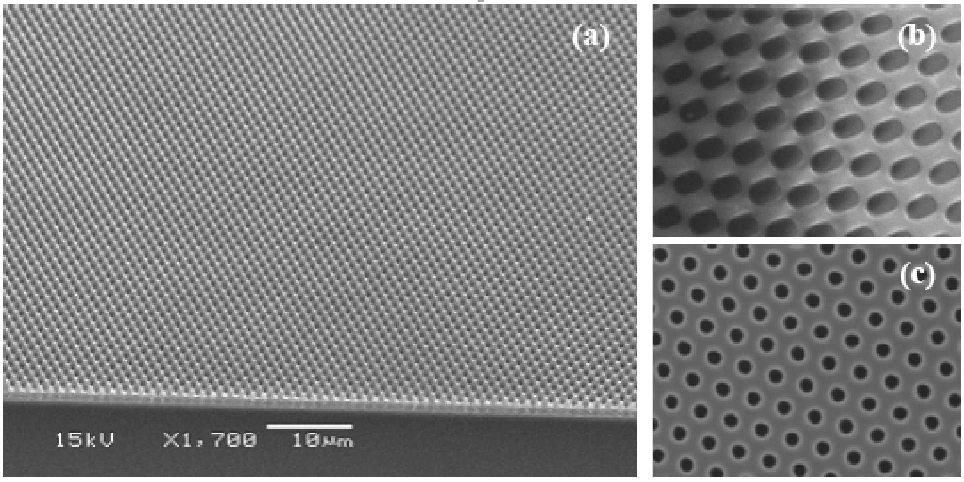
Laser direct-writing is widely used in materials modification, micro-optics manufacturing, high-density 3D optical storage manufacturing and so on. However, the throughput and repeatability are low with relatively high cost. It is difficult to fabricate 3D nanostructures inside non-photosensitive materials.

2.2 Laser interference micro/nano manufacturing

Laser interference is promising for low-cost and large-area manufacturing of sub-micro or nanoscale structures and devices, which does not need exposure, development, setting, and etching (Kawamura et al., 2001).

Lai et al. (2005) employed two-beam interference to fabricate 2D square, hexagonal structures, 3D rectangular-square, and hexagonal-hexagonal structures on SU-8 photoresist as shown in Figure 5. The periodic structures are very uniform in a large area of 6 mm × 6 mm. Kondo et al. (2003) obtained rod structures in square lattice by using multiphoton-absorption four beams interference, where the rod thickness and shape were controlled. Zhong et al. (2010) fabricated highly uniform gratings, quasicrystals and 3D fcc-type structure photonic crystals using two-photon holographic lithography. Lasagni et al. made 3D periodic pillars in a square lattice on PETIA by two-photon polymerisation with three, four and five-beam interference as shown in Figure 6 (Lasagni et al., 2009).

Figure 5 2D periodic structures, (a) square structure (b) hexagonal structure (c) hexagonal structure



Source: Lai et al. (2005)

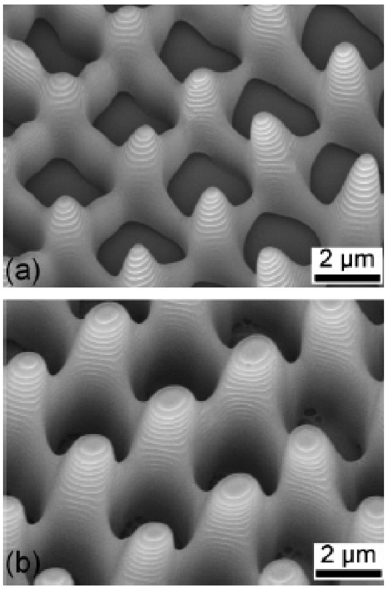
Future work is expected to investigate

1 how to control structure period, shape and size in laser interference manufacturing

2 how to fabricate high-resolution complex 3D and aperiodic microstructures.

Polarisation-dependent nanoscale periodic structure can be formed by a femtosecond laser pulse and its induced plasma, which is also an important research direction of femtosecond laser interference manufacturing.

Figure 6 3D periodic pillars fabricated by two-photon polymerisation using five-beam interference

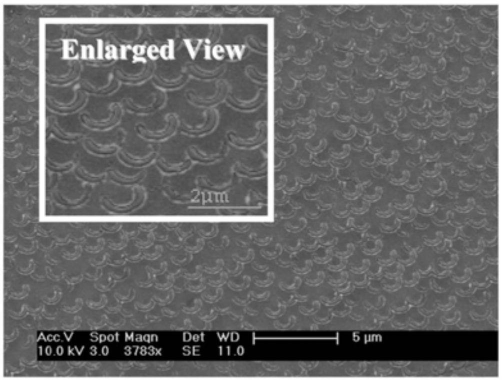


Source: Lasagni et al. (2009)

2.3 Laser near-field nanomanufacturing

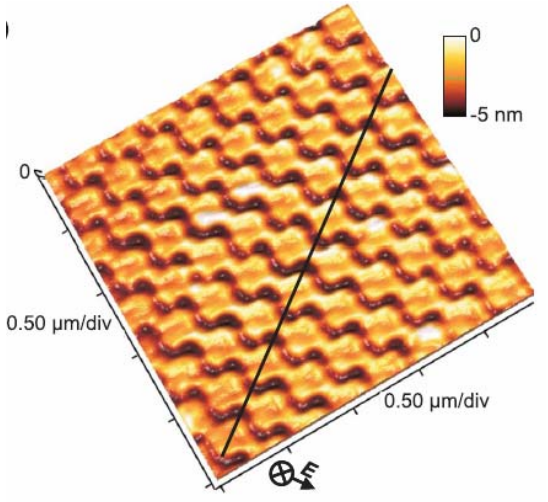
Laser near-field manufacturing is very promising for sub-diffraction-limit nanomanufacturing (Wang et al., 2010), which is typically implemented by combining a laser with scanning probe microscope (scanning tunnelling microscope, atomic force microscope, or scanning near field optical microscope) or special nanostructure. Gorbunov and Pompe created small hillocks with silver tips on the surface of gold films in vacuum (Gorbunov and Pompe, 1994). Then, structures with a lateral resolution of approximately 20 nm were achieved, for which various metal tips such as tungsten, silver, and platin/iridium tips can be used (Jersch et al., 1996). In additional to 3D nanostructures, nano-oxidation can be obtained by using laser near-field manufacturing. Lu et al. fabricated 20–30 nm nano-oxidation dots and 30-nm-width nano-oxidation lines on hydrogen-passivated Ge (100) surfaces by a nanosecond laser and STM (Lu et al., 1999), which may be due to locally enhanced optical field instead of thermal expansion of STM tip (Lu et al., 2000). Yi et al. (2008) made high-conductivity nanoscale dots and lines on silicon substrates and successfully controlled the nanostructure sizes by the number of laser pulses. However, with a resolution of 20 nm, STM-based near-field manufacturing is of poor controllability and repeatability with high requirements of environment and material conductivity. In contrast, AFM-based laser near-field manufacturing releases high requirements of environment and material conductivity, with a better resolution of 10 nm. Chimmalgi et al. produced various nanostructures including lines, grids and craters with 10 nm resolution by a femtosecond laser and AFM (Chimmalgi et al., 2003). Wang et al. (2006) obtained nanostructures of sub 50 nm resolution in positive photoresist by a 355 nm laser near field enhancement with bowtie apertures. Their work demonstrates that bowtie apertures are better in term of both near field enhancement and light concentration than square apertures and rectangular apertures.

Figure 7 An ordered array of c-shaped structures



Source: Guo et al. (2007)

Figure 8 AFM image of nanochannels (see online version for colours)



Source: Wang et al. (2008)

By using lens array, Guo et al. (2007) formed the locally enhanced optical field through self-assembled particle array instead of SPM tips and obtained 6 × 106 nanolines and c-shaped patterns of an 80 nm resolution in a large area of 5 mm × 5 mm (Figure 7). Hubenthal et al. (2009) made a 4-nm-depth, 96-nm-width and 6-μm-length nanochannels by nanoparticle arrays. Wang et al. (2008) fabricated nanoscale ring bumps and convex bumps on glass surface or inside glass by using a femtosecond laser and optical near-field effects in water (Figure 8). Moreover, in water, focal length is greatly extended due to the multiple focusing spots and nanostructure can be obtained inside the glass. The method makes it possible to produce multiple complex 3D nanofeatures simultaneously inside solids.

Self-assembled particle arrays may create 3D nanostructures simultaneously. But compound aperiodic structures cannot be achieved, on which further investigations can be focused. Also, laser near-field manufacturing is of very low throughput, poor controllability and poor repeatability. It remains a challenge to understand the mechanism of laser near-field manufacturing. STM, AFM or SNOM needs very sophisticated devices to control the distance between the tip and target surface. The future applications critically depend on the development of control technology of scanning probe microscope.

3 Applications

3.1 Laser manufacturing of nanoparticles

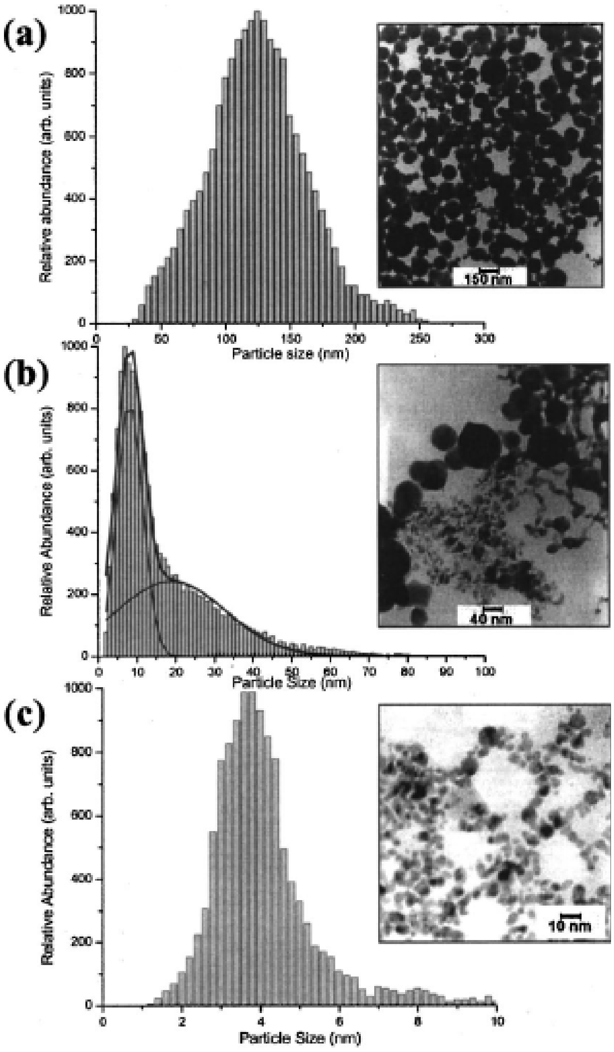
There are two main ways for lasers to fabricate nanoparticles:

1. Nanoparticles is produced on substrates by using laser ablation in vacuum or a gas. Liu et al. (2007) generated nickel nanoparticles by a femtosecond laser, in which compositions and structures could be selectively generated by controlling background gases.

2. Nanoparticle colloids are produced by laser ablation in a liquid. Kabashin and Meunier (2003) prepared colloidal gold nanoparticles in water by user a femtosecond laser and small and uniform nanoparticles were produced at low fluences, while large and broad-size nanoparticles were produced at high laser fluences. Sylvestre et al. (2004) proposed a novel method by using femtosecond laser ablation in cyclodextrins which could significantly improve the size uniformity and stability of the prepared colloidal metal nanoparticles.

Many studies have been focused on laser parameters and environment characteristics to unify and reduce nanoparticle sizes, for which the combination of mechanical, optical and chemical synthesis may be a good solution. Laser ablation was recently used for liquid metal nanoparticles generation, which deserves future research attentions. For example, how to select solution for desired nanoparticles? How to choose laser fluence and wavelength? How to improve productivity? How to model laser-liquid-nanoparticle interactions?

Figure 9 TEM images and size distributions of gold nanoparticles, (a) 1,000 J/cm^2 (b) 160 J/cm^2 (c) 60 J/cm^2

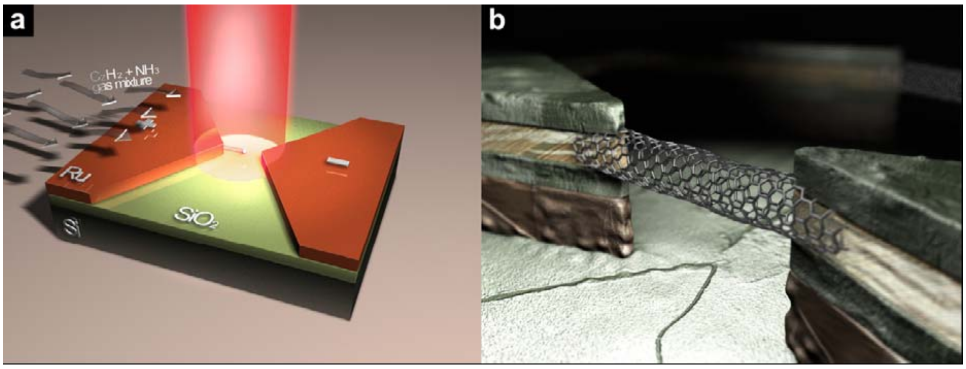


Source: Kabashin and Meunier (2003)

3.2 Laser manufacturing of nanotubes

Chemical vapour deposition (CVD), laser ablation and arc discharge can be used to fabricate carbon nanotubes with high throughput, but of low quality and high cost (Moors et al., 2009). In recent years, laser-assisted CVD (LCVD) has been developed as a novel technique with advantages of precise position and length/direction control of SWNTs growth at low substrate temperatures (Xiong et al., 2009; Zhou et al., 2010). LCVD was used to achieve in-situ growth and integration of SWNTs into pre-designed nano-architectures by the Lu's group (Figure 10). The SWNT-integrated structures were fabricated on p-type silicon substrates covered by a 2µm SiO2 layer for electrical insulation. Metallic electrode patterns containing multiple pairs of tip-shaped electrodes were fabricated after general photolithography and sputtering processes. Metallic electrode consisting of multiple metallic layers, including a 200 nm ruthenium (Ru) layer and Al/Fe/Al (5/1/20 nm) films, were deposited on a Ru film as a bimetallic catalyst to grow SWNTs. A continuous-wave (CW) CO2 laser was focused on the tips of the electrodes on the patterned substrates to provide localised field which initialised SWNTs growth at the sharp tips with Fe catalysing. A DC voltage was applied between the electrodes to direct the growth direction of SWNTs. SWNT bridges SEM (Figure 11) showed a SWNT connecting the cathode and the anode with the diameter of about 12 nm.

Figure 10 (a) Schematic diagram of the LCVD manufacturing process and (b) schematic diagram of an SWNT-integrated bridge structure (see online version for colours)



Source: Zhou et al. (2010)

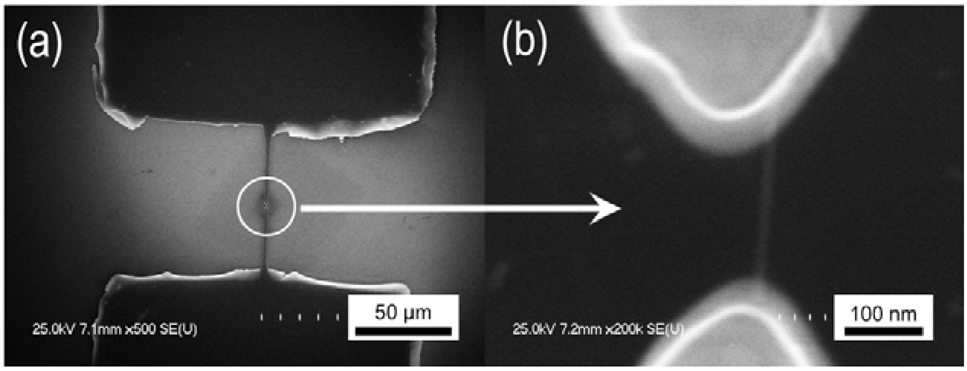
Three important research directions of laser manufacturing of nanotubes include

1 How to obtain desired radius, longer length and higher purity?

2 How to effectively and efficiently control growth positions and directions?

3 How to realise large-scale CNTS synthesis with uniform high qualities?

Figure 11 SEM of (a) the electrodes and (b) the SWNT-bridge structure

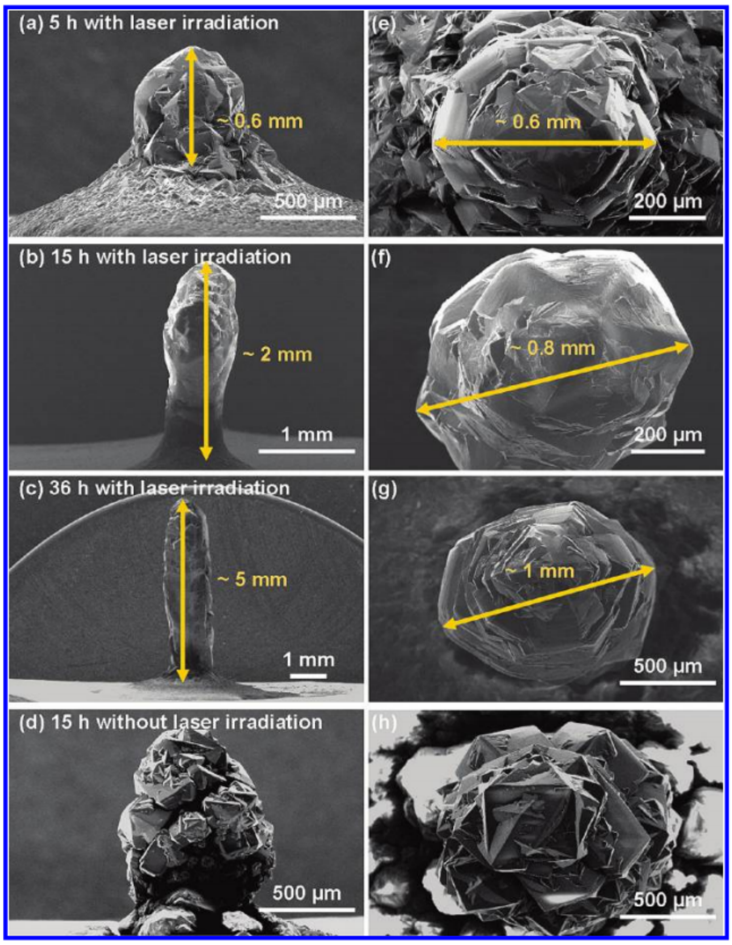


Source: Xiong et al. (2009)

3.3 Laser assisted crystal manufacturing

Many techniques, including CVD, pulse laser deposition (PLD), and shock wave compression technique have been used to synthesise diamond crystals on various substrates (Novotny et al., 2004), which typically requires high temperature and high-pressure, and are typically based on the reactions in a vacuum chamber.

Figure 12 SEM images of diamond crystals growth with laser excitation at 10.532 μm (see online version for colours)



Source: Xie et al. (2010)

High-quality diamond crystals with high growth rates is produced in open air (Xie et al., 2010) by using laser assisted CVD method. Ethylene (C_2H_4) was introduced into the oxyacetylene combustion flame to couple the laser energy into the combustion reaction through the resonant excitation of the ethylene molecules. Diamond crystals with a length of 5 mm and a diameter of 1 mm were obtained on silicon substrates as shown in Figure 12. The resonant excitation was utlised for selective bond breaking and reaction pathways control. It was demonstrated that both diamond growth rate and film quality can be improved by using resonant laser coupling (Ling et al., 2009a, 2009b). Laser resonance absorption may be an important future way to improve the controllability, productivity and selectivity of micro/nano-fabrication.

4 Conclusions

Laser, especially femtosecond laser, micro/nano fabrication is an active interdisciplinary frontier, which involves manufacturing, optics, physics, chemistry, and materials. It presents unique advantages of 3D subwavelength fabrication with great application potentials in automotive industry, pharmaceutical industry, process and automation technology, defence industry, aerospace industry, information technology, telecommunication technology, biotechnology, medicine industry, measurement and microscopy, environmental technology.

An integrated multiscale physico-chemical modelling shall be established to understand the ultrashort, non-equilibrium and non-linear laser-materials interactions from nanometre to millimetre and from femtosecond to microsecond. Mechanisms of material removals by femtosecond lasers remain controversial. The particular mechanism strongly depends on material properties, laser fluence, pulse duration, wavelength, and number of pulses. However, it remains a challenge to clearly explain the transition from one phase change mechanism to another and predict the roles of the competing ablation mechanisms.

With increasing powers, in future, femtosecond laser direct writing may achieve high-speed, low-cost mass production of high-quality devices. Femtosecond laser interference does not need exposure, development, setting, and etching and can form periodic functional structure on/inside transparent materials. Future work is expected to investigate

1. how to control the period, shape and size of the structures fabricated laser interference manufacturing
2. how to fabricate high-resolution complex 3D and aperiodic microstructures.

Polarisation-dependent nanoscale periodic structure can be formed by a femtosecond laser pulse and its induced plasma, which is also an important research direction of femtosecond laser interference manufacturing.

Laser near-field manufacturing is very promising. But, it is of very low throughput, poor controllability, and poor repeatability. It remains a challenge to understand the mechanism of laser near-field manufacturing. STM, AFM or SNOM needs very sophisticated devices to control the distance of near field. The future applications critically depend on the development of control technology of scanning probe microscope.

Many studies have been focused on laser parameters and environment characteristics to uniform and reduce nanoparticle sizes, for which, the combination of mechanical, optical and chemical synthesis may be a good solution. Laser ablation was recently used for liquid metal nanoparticle generation, which deserves future research attentions. For example, how to select solution for desired nanoparticles? How to choose laser fluence and wavelength? How to improve productivity? How to model laser-liquid-nanoparticle interactions?

Three important research directions of laser manufacturing of nanotubes include

1. How to obtain desired radius, longer length and higher purity?
2. How to effectively and efficiently control growth positions and directions?
3. How to realise large-scale CNTS synthesis with uniform high qualities?

Laser assisted CVD method was adopted to produce high-quality crystals with high growth rates in open air. Laser resonance absorption may be an important future way to improve the controllability, productivity and selectivity of micro/nano-fabrication, with some important questions, for example:

1. How to design multiple resonance absorptions in a fabrication process based on the understanding of electron dynamics in multiple laser fields?
2. Is it possible to control/changes chemical reactions/phase change mechanisms during the fabrication?
3. How to achieve selective fabrication in terms of materials or even chemical bands?

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