Nano-Sized and Periodic Structures Generated by Interfering Femtosecond Laser

Yoshiki NAKATA*, Kunio TSUCHIDA*, Noriaki MIYANAGA*, Tatsuo OKADA**

*Institute of Laser Engineering (ILE), Osaka University, 2-6 Yamadaoka, Suita-shi, Osaka 565-0871, Japan
nakata-y@ile.osaka-u.ac.jp
**Graduate School of Information Science and Electrical Engineering (ISEE) , Kyushu University, 744 Motooka, Nishi-ku, Fukuoka-shi, Fukuoka 819-0395, Japan

Top down technique of interfering femtosecond laser processing was applied to generate new nanomaterials such as nanocrown, nanomesh, nanobelt, and dual periodic structure. In the case of nanocrown, whiskers are standing at the edge of a nanohole. The width of a whisker was just 80 nm. A nanohole array structure generated by interfering 4 beams was applied to a spatial filter as a base material for the growth of photo-polymerizing polymer, and polymer nanobump array was generated. Dual periodic structures were generated by multiple shots, and bimetallic nanobelt was generated from bilayer thin film. Processing of fiber film was tested.

DOI: 10.2961/jlmn.2008.02.0001

Keywords: Interfering femtosecond laser, nanocrown, polymer nanobump, photo mask, bimetallic nanobelt, dual periodic structure

1. Introduction

Nanotechnology based on nano-textured surface or nanomaterials such as nanowire, nanotube and nanobelt is a topic for many years. They have been produced by bottom-up techniques, but they have difficulties in versatility, size and structure control, and alignment. On the other hand, material processing using interfering femtosecond laser beams was applied to generate nano-sized and periodic structures on and in materials [1-10]. In addition, we have been generated new nano-sized structures by thin film processing using interfering femtosecond laser beams [1-7]. In this paper, our recent results of the generation of nanomaterials and nano-textured surfaces are reported.

2. Experimental setup[5]

In this experiment, interfering femtosecond laser beams are used for material processing, so a beam correlator is requirement. A general beam correlator is composed of a mirror beam splitter and mirrors as shown in Fig. 1 (a). On the other hand, a set of transmission beam splitter and Fourier optical system can be used as a beam correlator, as shown in Fig. 1 (b). In this case, a beam is split by the transmission beam splitter, and then the split beams are correlated by the two convex lenses. The advantages of the latter system is as followings; the setup is simple, the number of beams can be changed by changing the transmission beam splitter and dumper, the optical delay is automatically adjusted, and even mask transfer can be achieved [2, 3]. The most important advantage is limitless interfering area as shown in the past paper [7], though it is limited to some hundreds micron in the case of the system in Fig. 1 (a). In this case, the wave planes correlate with an angle, and the correlated region is restricted. On the other hand, in the case of the system in Fig. 1 (b), they correlate without an angle, so there is no geometric restriction [5]. The idea to use a Fourier optical system as a beam correlator had been proposed by Maznev et al. in 1998 [11]. In their experiment, they used the system only for second harmonic generation. Here, higher femtosecond laser fluence is required to achieve laser processing, so we have proposed two schemes for beam correlator which can be used for laser processing, as shown in Fig. 1 (c) and (d). The former is demagnification system, and the latter is cylindrical lens system. In the case of demagnification system, focal length of L2 is shorter than that of L1, so the original beam shape is demagnified as shown in the inset of Fig. 1 (c), and the fluence is increased. On the other hand, in the case of cylindrical lens system, the original beam shape is additionally narrowed as shown in the inset of Fig. 1 (d), and fluence is increased. This system is useful to generate periodic structure inside of a transparent material [6]. In both systems, the period of the interference pattern will be $L = M\Lambda/2$, where Λ is the period of the transmission grating, $M = f2/f1$ is the magnification factor of the system. To show the advantage of our system about correlating region, gold thin film was processed using the cylindrical system. A line of periodic holes structure with 6 mm length could be generated in a single shot, as shown in the past paper [7]. The maximum fluence is limited by the damage threshold of the lens *L2* in the case of the demagnification system, but is not limited in the case of the cylindrical lens system. In addition, a Schwarzschild optics can be used as a beam correlator as shown in Fig. 1 (e), but the beam diameter is restricted in the size of the window, and 0^{th} order beam can not be used. Comparing the systems in Fig. 1, the demagnification system and the cylindrical lens system are the most available systems.

In this experiment, an ultrashort pulse laser system, of which the pulse width was 120 fs to some ps and the center wavelength was about 800 nm, was used. Monolayer or bilayer thin films at room temperature were processed without any evacuation.

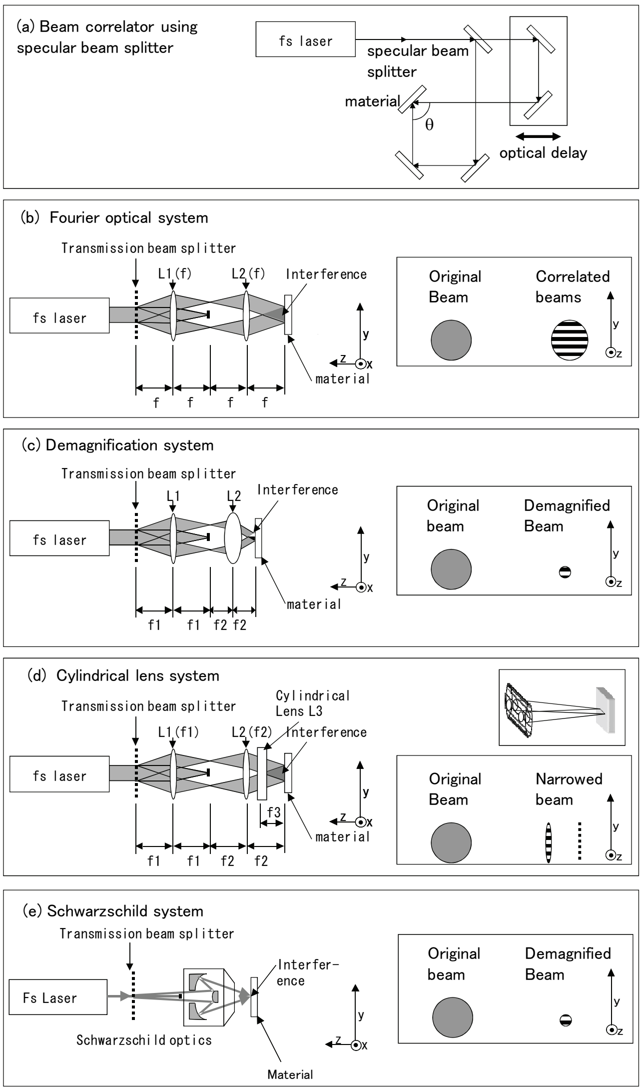

Fig. 1 Beam correlators using (a) specular beam splitter, (b) Fourier optical system, (c) demagnification system, (d) cylindrical lens system, and (e) Schwarzschild optics.

3. Results

3.1 Nanocrown array structure

By changing the target structure, the basic structure can be also changed. Here, nanocrown array structure was firstly generated as shown in Fig. 2. The target material was a bi-layer thin film of 50 nm thick Au-Sb on 5 nm thick Cr on Silicon substrate. Interfering 4 beams was also used, and the pulse width was about 120 fs. In a single shot, periodic nano-sized crown-like structure could be generated as shown in the bird's eye view. It seems that whiskers are standing on the edge of each hole. The height was about 400 nm, and the width was just about 80 nm. Here, in the case of Au thin film on silica glass substrate, no whisker was generated as shown in the past paper [4], and in Fig. 2 (b). There are some different parameters between these two processes such as film property, thermal conductivity and optical property of the substrate, correlation angle. Detailed experiment is required to know the key parameter for the generation of the crown-like structure.

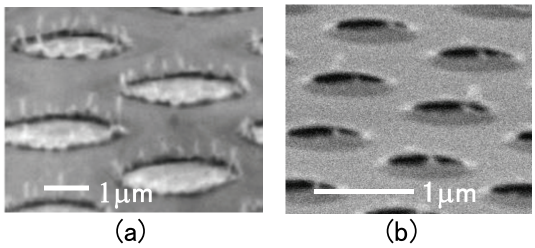

Fig. 2 (a) nanocrown array structure and (b) nanohole array structure.

3.2 Nanohole array as a photo-mask

Nano-sized and periodic structure is quite useful for the base material of the growth of another nanostructure. Here, a nanohole array was used as a mask for the generation of nanobump array of polymer. The procedure is explained in Fig. 3. (a) a nanohole array is used as a template. (b) photo-polymerizing polymer (Norland NOA 81) immerses the template. (c) The sample was irradiated from the backside, and solidification occurs. (d) After washing by ethanol, nanobump array of polymer can be generated. This technique is useful to generate nano-sized and periodic polymer nanobump array with low photon cost compared to the photo-polymerizing process using femtosecond laser [9].

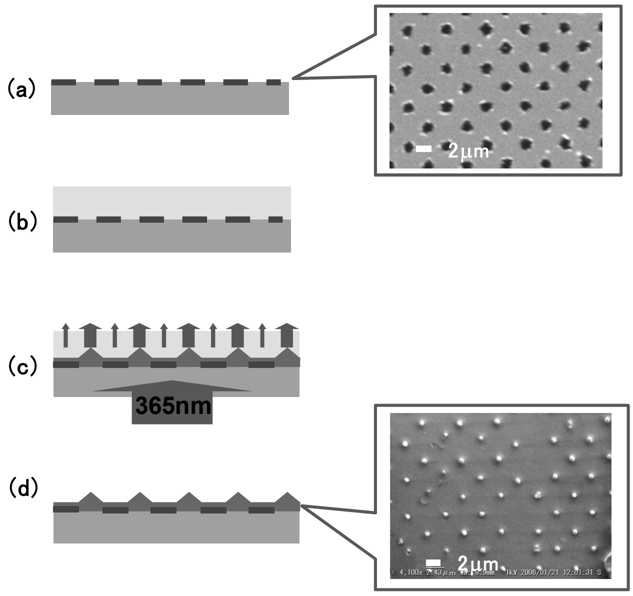

Fig. 3 Nanohole array mask as a template for the generation of nanobump array by photo-polymerization.

3.3 Bimetallic nanobelt structure

Our technique is top-down technique, so any thin film can be used as raw materials. The adaptability is far better compared to any bottom-up techniques such as CVD, discharge, anodization, etc.. Here bimetallic nanobelts were generated as a demonstration, as shown in Fig. 4. A bilayer thin film on silica glass substrate, of which the upper layer was 350 nm thick aluminum and the lower layer was 200 nm thick gold, was used as a raw sample. It was ablated by interfering 2 beams, and grating structure was generated. By exfoliating it, bimetal nanobelts with 550nm thickness cold be generated as shown in Fig. 4. Such nanobelt structure can never be generated by any bottom-up techniques.

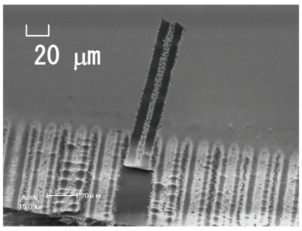

Fig. 4 Gold-aluminum bimetal nanobelt structure.

3.4 Dual periodic structure

It is known that quasi-periodic structure can be generated by multi laser shots [13]. By using multi shots of interfering beams, dual periodic structure can be generated as shown in Fig. 5. Here, Cr thin film with 44 nm deposited on a silica glass was used. The number of shot was 25, and the fluence was about 54 mJ/cm^2. The longer periodic structure with 1.2 μm period is due to the interference, and shorter quasi-periodic structure is due to the multi shots. The period is 160 nm in average, and the width of each structure is 74 nm. The width of the processed region is about 530 nm. The quasi-periodic structure is perpendicular to the polarization of the laser, as in the case of singe beam processing. White flocculates are thought to be condensed Cr. Such special periodic structure will have complex surface functionality, such as coefficient of friction or hydrophilicity.

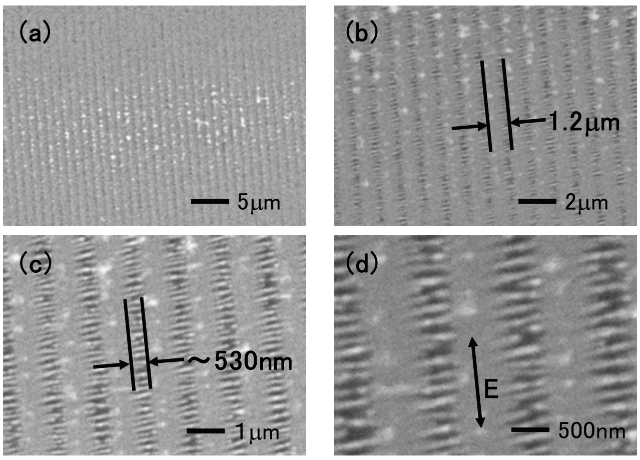

Fig. 5 Dual periodic structure generated on Cr by multi shots of 2 interfering beams. Each picture shows the same structure observed at different magnification.

On the other hand, it is interesting that dual periodic structure can be generated without interference as shown in Fig. 6. The number of shots was about 150. The mechanism could be the combination of the interference of plasmonic wave and standing sound wave, etc. Detailed investigations are required to know the mechanism.

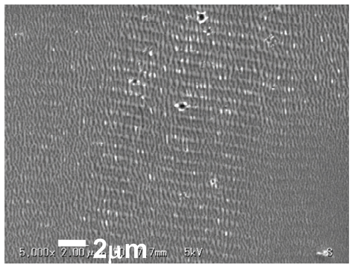

Fig. 6 Dual periodic structure generated on Al by multi shots of single beam.

3.5 Processing of carbon fiber

Fiber carbon is known as a light and hard material, and has been used as frames, sheets, etc.. These days it is also used in nanotechnology, for example FED electrode. There are needs to process them in higher-resolution, so a sample film was processed by multi shots of femtosecond laser.

A beam was focused on the surface with f = 60 mm lens, and the pulse energy was about 0.8 mJ. The fibers were cut and the surface was modified at the spot by 6 shots, as shown in Fig. 7 (a). At higher magnification as shown Fig. 7 (b), micron and random quasi-periodic structure can be seen. On the other hand, the hole was deep , and annual-ring like structure with shorter period could be generated as shown in Fig. 7 (c) and (d). The period was around 200 nm from top view. It is thought that the origin of the structure is the intrinsic structure of fiber, because the structure has different direction at different points, and is not related to the electrical field as shown in Fig. 7.

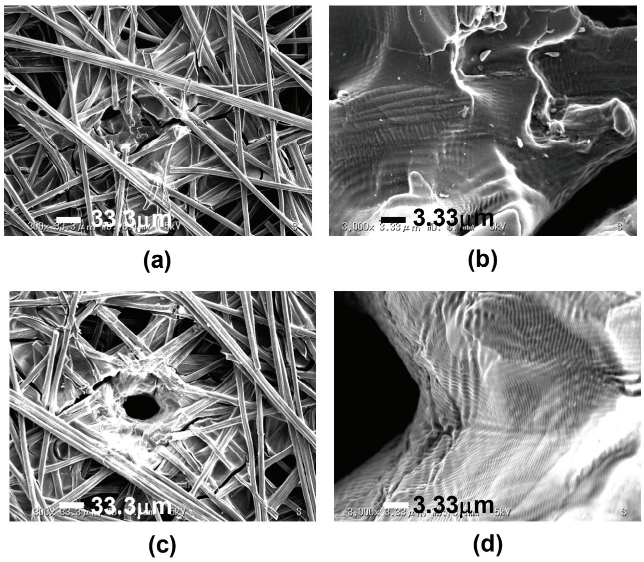

Fig. 7 Quasi periodic structure generated on carbon fiber film. The number of shot were (a) (b) 6 and (c) (d) 25, respectively.

4. Summary

A variety of nanostructures such as nanocrown, polymer nanobump, bimetallic nanobelt, and dual periodic structure could be generated by interfering femtosecond laser processing. This technique is top-down technique, and different from the bottom-up techniques such as CVD and plasma process as follows; the bottom-up techniques can produce some nm-sized structures such as nanowire, nanotube and nanoparticles. On the other hand, some tens or hundreds nm sized structures can be generated by our technique. The structures are quite unique and controllable, and aligned structure can be generated automatically in our case. The adaptability to different material is very good because of top-down technique. In practical point of view, no special ambient such as high vacuum, temperature control, special gas or liquid is required. This method will improve the existing applications and new opportunities of nano-sized structures.

Acknowledgments

Some experiments were performed by the femtosecond laser at Venture Business Laboratory and Graduate School of Science at Osaka university, and the Institute for Ionized Gas and Laser Research, Kyushu university. This research was partly supported by Japan Science and Technology Agency, Japan.

References

[1] Y. Nakata, N. Miyanaga, T. Okada: Appl. Surf. Sci., **253**, (2007) 6555.
[2] Y. Nakata, T. Okada, and M. Maeda: Appl. Phys. A, **79**, (2004) 1481.
[3] Y. Nakata, T. Okada, and M. Maeda: Opt. Las. Eng., **42**, (2004) 389.
[4] Y. Nakata, T. Okada, and M. Maeda: Jpn. J. Appl. Phys., **42**, (2003) L1452.
[5] Y. Nakata, T. Okada, and M. Maeda: Appl. Phys. A, **77**, (2003) 399.
[6] Y. Nakata, T. Okada, and M. Maeda: Jpn. J. Appl. Phys., **42**, (2003) L379.
[7] Y. Nakata, T. Okada, and M. Maeda: Appl. Phys. Lett., **81**, (2002) 4239.
[8] K. Kawamura, M. Hirano, T. Kamiya and H. Hosono: J. NonCryst. Sol. **352** (2006) 2347.
[9] T. Kondo, S. Juodkazis, V. Mizeikis, S. Matsuo and H. Misawa: New Journal of Phys. **8** (2006) 250.
[10] J.H. Klein-Wiele, J. Bekesi, P. Simon, Appl. Phys. A **79**, (2004) 775.
[11] A.A. Maznev, T.F. Crimins, K.A. Nelson, Opt. Lett., **23**, (1998) 1378.
[12] P. Baumgart, D. J. Krajnovich, T. A. Nguyen, and A. C. Tam: IEEE Trans. Magn., **31**, (1995) 2946.
[13] J. Heitz, E. Arenholz, D. Bäuerle, R. Sauerbrey, H. M. Phillips: Appl. Phys. A, **59**, (1994) 289.

(Received: May 28, 2007, Accepted: January 15, 2008)