Topdown femtosecond laser-interference technique for the generation of new nanostructures

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Abstract. Precise and periodic structure can be formed on and inside materials by interfering femtosecond laser processing. In this method, laser ablation or modification is induced according to the interference pattern. In our technique, this method is applied to thin film processing, and nano-sized structures such as nanobumps, nanomeshs, and nanobelts. Nanobump array structure can be formed by periodic thermal processes induced by a single shot of interfering laser. Field emission from a nanobump array was demonstrated. By processing at relatively higher fluence, a grating, a circular or an ellipsoidal hole array structure can be generated according to the number of beams. By exfoliating the structures, nanobelt or nanomesh structure can be generated. This is a top-down technique for the generation of new nanomaterials.

1. Introduction
A variety of nanomaterials have been produced by bottom-up technology such as plasma process, chemical process, etc. However, these processes have difficulty in adaptability to different materials, size and structure control, and alignment. On the other hand, material processing by femtosecond (fs) lasers has been receiving great interest for a decade. By using this method, a material can be processed with a resolution of wavelength range. Moreover, material processing using interfering fs laser was applied to generate nano-sized and periodic structures on and inside materials in a single process [1-3]. Recently, we generated new nanostructures from thin film processed by interfering fs laser beams [4-7]. In this technique, ablation and thermal modification are effectively applied to generate special structures. In this paper, we review our recent results of the generation of new nanostructures such as nanobump arrays, nanomeshs and nanobelts.

2. Experimental Setup
The experimental setup was shown in past papers [5,6]. An ultrashort pulse laser, of which the pulse width was about 100 fs and the center wavelength was about 800 nm, was used. A demagnification optical system or a cylindrical lens system was used as a beam correlator. The period of the processed structure can be calculated by \( L = MA/2 \) when first-order diffraction beams are used, where \( A \) is the
period of the transmission grating, and \( M \) is the magnification factor. Gold and bi-layered thin films were used as samples. All the processes were done in ambient air at room temperature.

3. Experimental Results

3.1.1. Nanobump structure

In a past paper, we have reported that a hollow nanobump array structure can be generated on a metallic thin film in a single shot of interfering fs laser beams [4]. The shape could be manipulated to conical, bead on bump, standing bead on hole, ellipsoidal and linear according to the applied fluence and the number of interfering beams. Here, the structure change of conical nanobump arrays as a function of pulse width is reported. The number of beams was four, and the fluence was about 87 mJ/cm². Laser structuring was made on gold films, usually of 50-nm thickness evaporated on silica glass.

Figure 1 shows the nanobump arrays generated at different pulse widths. When the pulse width was 120 fs, conical nanobumps were generated as shown in figure 1 (a). The aspect ratio between height and width was about 0.6, which is two orders of magnitude larger than that generated by typical laser texturing techniques, for example, the formation of forming bumps on metals with a nanosecond laser [8]. On the other hand, when the pulse width was about 1.2 ps, smaller nanobumps were generated as shown in Figure 1 (b). The decrease of the size with the increase of the pulse duration might be due to the energy loss by electron diffusion and thermal diffusion.

Nano-sized structures are useful for field emission due to field enhancement effects. Here, field emission from a nanobump array was demonstrated. Nanobump samples with 200 nm or smaller height were used as a cathode and negatively biased. An aluminum plate was used for the anode, and was grounded via a resistance of 47 kΩ. The distance between the two electrodes was 50 μm. The degree of vacuum was about 10⁻³ Pa. The field emission current density as a function of applied field is plotted in figure 2 (a). Field emission appeared at 10.6 V/μm. The turn-on field, namely the electric field required to produce a current of 0.01 mA/cm², was 13.6 V/μm. In the case without nanobump, no field emission was observed at these electric fields. The Fowler-Nordheim (FN) theory, which describes field emission, gives a straight line response in the so-called FN plot. Figure 2 (b) is the corresponding FN plot where two lines can be seen that are typical in field emission phenomena.

![Figure 1. Nanobump arrays generated at (a) 120 fs and (b) 1.2 ps. The top sketch depicts the multi-beam orientation.](image_url)
3.1.2. Nanobelt structure

The combination of fs processing and peeling is useful to generate new nanostructures [7]. Nanobelt structure can be generated by thin film processing with two interfering fs beams, as shown in figure 3. The gold film thickness was 10 nm. By increasing the laser fluence to about 54 mJ/cm², the ablated region was widened and thin nanobelts were formed with 200 nm width as shown in figure 3 (a). The size is comparable to that generated by the CVD method [9]. Moreover, a bimetallic nanobelt was generated from bi-layered thin films evaporated on silica glass as shown in figure 3 (b). The lower layer was 200 nm thick gold, and the upper layer was 350 nm thick aluminum. The film was ablated by two interfering fs beams by using a cylindrical lens system [5], and the film was scanned parallel to the interfering pattern during the process. Standing two bimetallic nanobelts were exfoliated from the substrate by scratching. The length was over 100 μm, as shown in figure 3 (b). This experiment demonstrates the adaptability to different materials of this technique, since such structure can not be generated by the CVD method. The bimetallic nanobelt is useful for fiber, actuator and sensor applications.

3.1.3. Nanomesh structure

By ablating thin films with four or three interfering beams, nanomesh can be generated as shown in figure 4. In the case of four beam interference, circular holes were generated. The diameter was 680 nm at the fluence of 19 mJ/cm², and 1.4 μm at 54 mJ/cm² as shown in figure 4 (a) and (b), respectively. On the other hand, ellipsoidal holes were generated in the case of three beam interference as shown in figure 4 (c). The aspect ratio was 1.8.
4. Summary
A variety of nanostructures were generated by thin film processing with interfering fs laser beams. The advantages of this technique are as follows: adaptability to different materials, size and structure control, alignment control, and no requirement for special ambient such as high vacuum and temperature control. This method will improve existing laser applications and offer new opportunities in nanotechnology and laser technology.

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Figure 4. SEM images of nanomesh generated with (a) 19 mJ/cm² fluence with four beams, (b) 54 mJ/cm² fluence with four beams, and (c) 41 mJ/cm² fluence with three beams. The top sketches depict the beam orientation.