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Multiphoton fabrication of periodic structures by multibeam interference of femtosecond pulses

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Femtosecond laser pulses are useful for laser microfabrication through multiphoton absorption. However, it is difficult to create interference of femtosecond pulses for the fabrication of periodic structures. In this letter, we report the fabrication of two-dimensional periodic structures by means of multibeam interference of femtosecond pulses. Scanning electron microscopy revealed a rod structure arranged into a square lattice. The possibility of controlling the period of the lattice, rod thickness, and rod shape were demonstrated. © *2003 American Institute of Physics.*
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Thanks to the recent developments in ultrafast laser technology, the field of femtosecond (fs) laser processing is seeing wider application.[1,2] In particular, fs laser processing has been successfully applied to three-dimensional (3D) microfabrication. By a tightly focused fs pulse, multiphoton absorption (MPA) is induced in a region smaller than the diffraction limit in both the lateral and axial directions. This allows finer 3D structures in photopolymers and transparent solids.[3,4]

One of the significant applications of fs laser processing is the fabrication of photonic crystals (PhCs). A PhC is an artificial, multidimensional periodic structure whose period is in the order of the wavelength of light.[5–7] PhCs are attracting interest due to their promising applications in, for example, integrated optical circuits and low-threshold lasers. Development of a practical fabrication technology is one of the most important tasks in the field of PhC research. Laser microfabrication of PhCs have been reported by several groups.[8–12] In these cases, a laser beam (or sample material) was scanned along a predesigned periodic pattern, and a PhC structure was constructed.

Another laser method of fabricating PhCs is laser beam interference. Laser interference creates a periodic modulation in electromagnetic field intensity. By transferring this intensity to a photosensitive material, a periodic structure is obtained. Fabrication of PhCs by laser interference has been carried out using cw,[13,14] nanosecond,[15] and fs[16] lasers. All of these reports used a one-photon absorption process. Use of a MPA process for laser interference would be also preferable in the fabrication of PhCs, for which fs pulses will be required. Recently, several reports have been published on the fabrication of gratings (one-dimensional periodic structures) by the interference of two fs pulses using MPA.[17–20] However, for structures with higher dimensional periodicity [two-dimensional (2D) or 3D], which are more interesting as PhCs, only our preliminary result has been reported.[21] This is, most probably, due to the difficulty in achieving the temporal overlap of at least three fs pulses, which is necessary for 2D periodic structures.[22]

The interference method, which uses a diffractive beam splitter (DBS), is useful for multibeam interference of fs pulses, since the temporal overlap of fs pulses is easily achieved.[16,23] Possible structures and their transmission spectra have been described theoretically in our earlier work.[21] In the present letter, we show 2D periodic structures fabricated using MPA in several experimental conditions, and demonstrate the possibility of controlling the structural parameters.

The optical setup used for the present experiments is shown in Fig. 1. Briefly, a DBS (G1023A or G1025A; MEMS Optical Inc.) divides the input laser beam into several, and the beams are collected on the sample by two

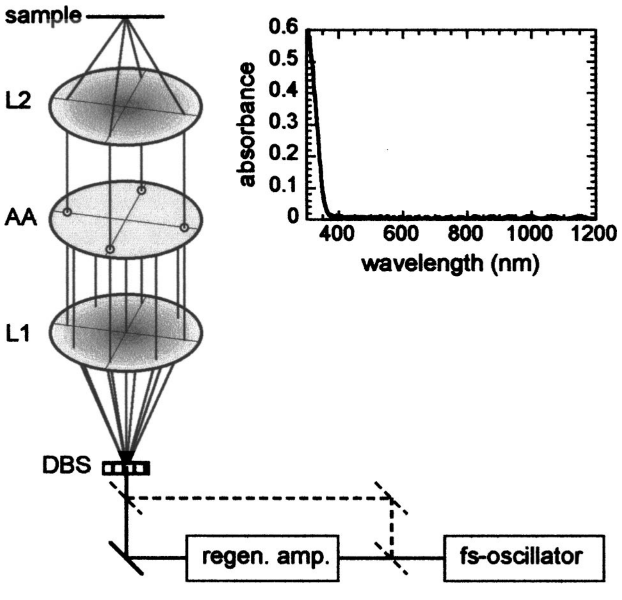

FIG. 1. Optical setup. DBS: diffractive beam splitter, L1 and L2: lenses, AA: aperture array. The inset shows the absorption spectrum of 4-μm-thick SU-8 film spin-coated on a coverglass.

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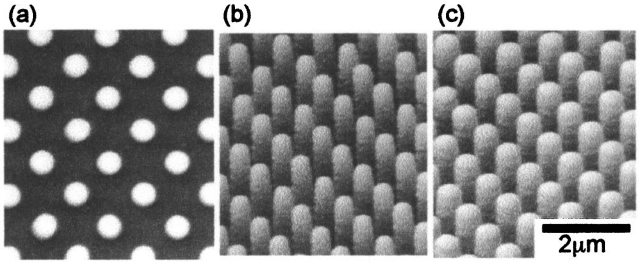

FIG. 2. SEM images of the structure fabricated by the four-beam interference of fs pulses. (a) Top view, (b) oblique view of the same sample, and (c) oblique view of another sample fabricated at larger exposure energy.

lenses. Temporal overlap of the divided pulses is achieved without adjusting the optical path lengths. Each beam was made to be parallel or slightly focused by the adjustment of the distance between the two lenses. Slight focusing increased laser power density and helped to make the MPA efficient. The beams meant to form interference were selected by an aperture array, which is placed between the two lenses. In the present study, only three or four beams (four equivalent edge lines of a quadrangular pyramid) were selected and used to create interference. In such cases, the fabricated structure will be homogeneous in the axial (z) direction, since the z component of wave vectors of the four beams are the same, and the periodicity of the structure is 2D. The interference angles θ_{air} (the angle between the main optical axis and the other beams in air) applied in the present experiments were measured to be 33.6°, 21.9°, and 10.8°.

Negative photoresist SU-8 (Microlithography Chemical Corp.) was used as an initial material for the fabrication. The absorption spectrum of SU-8 as shown in the inset of Fig. 1 indicates that one-photon absorption is negligible at an 800-nm wavelength. Consequently, it is expected that photopolymerization, if occurring, is due to a multiphoton reaction. The layer of SU-8 was spin-coated on a coverglass plate having a thickness of about 4 μm, and prebaked before exposure to fs pulses. After exposure, the sample was postbaked to enhance the photo-initiated crosslinking reaction. By the development after post-baking, unexposed regions were washed out, and a periodic structure was obtained. The structures were observed by a scanning electron microscope (SEM; Hitachi S-4200SE) after deposition of thin Au film by sputtering.

Fs pulses from a Ti:sapphire regenerative amplifier (wavelength of 800 nm, pulse duration of 150 fs, repetition rate of 1 kHz) were used for experiments. For comparison, pulses from a fs Ti:sapphire oscillator (wavelength of 800 nm, pulse duration of 80 fs, repetition rate of 82 MHz) were also used in some cases. The large difference in peak power ($\geqslant 10^4$ difference, while the average power is the same) enabled us to examine the multiphoton nature of the fabrication.

By using this method, periodic structures were fabricated. Periodic structures fabricated with an interference angle of 33.6° are presented in Fig. 2. In this figure, (a) and (b) show SEM images of the same sample from different perspectives. The single-pulse energy (sum of all beams) was $E_{\mathrm{tp}} = 16$ μJ and the exposure time was 60 s. The top view of the sample (a) reveals the periodic structure of a square lattice having a period of about 1.1 μm. The oblique view shown in (b) clearly demonstrates that the periodic structure consists of high-aspect-ratio rods. The height and diameter of the rods are about 4 and 0.6 μm, respectively; thus, an aspect ratio of about 7 was achieved. It should be noted that the height of the obtained structure is not limited by the coherent length of the pulse. A higher structure could be obtained if the stiffness of the material allowed the rods to withstand capillary forces during the development procedure. Figure 2(c) shows an oblique view of structures fabricated with a larger energy ($E_{\mathrm{tp}} = 26$ μJ, exposure time of 30 s). As seen, the lattice is the same as that in Fig. 2(b), but each rod is thicker. This difference demonstrates the possibility of controlling feature size by varying exposure conditions.

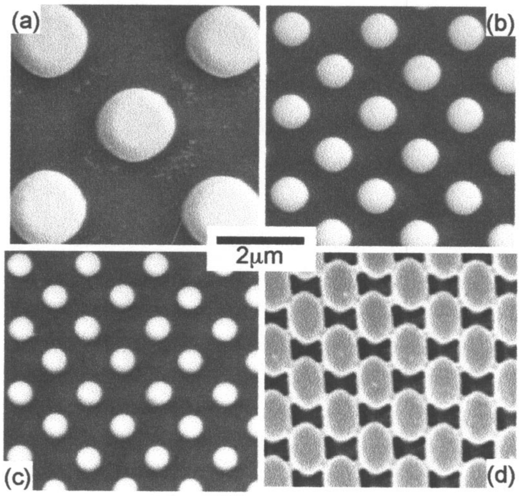

FIG. 3. (a)–(c): Top views of the fabricated structures by four-beam interference of fs pulses with interference angle (a) 10.8°, (b) 21.9°, (c) and (d) 33.6°. In (d) the number of interfering beams is reduced to three.

In order to examine the multiphoton nature of this photofabrication, we carried out exposure experiments using the much lower peak power pulses from the fs oscillator. Exposure with a large average power (130 mW) for a long period (1 h) was applied; however, no solidified structure was obtained after development. This result shows that MPA is responsible for the fabrication of the periodic structures in the present study.

Theoretically, intensity distribution $I(\mathbf{r})$ is calculated as

$$I(\mathbf{r}) = \left\langle \left| \sum_i E_i(\mathbf{r}) \right|^2 \right\rangle, \tag{1}$$

where

$$E_i(\mathbf{r}) = E_i^0 \cos(\mathbf{k}_i \cdot \mathbf{r} - \omega t + \phi_i) \tag{2}$$

is the electric field of each interference beam, and E_i^0 and ϕ_i are the electric field strength and the optical phase of the ith beam, respectively. If we make the simple assumptions that $E_i^0 = \mathrm{const}$ and $\phi_i = 0$, the structure in the x–y plane is a square lattice, in accordance with the experimental results. The period of the lattice calculated from the wavelength λ and the measured interference angle θ_{air} is $\lambda/(\sqrt{2}\sin\theta_{\mathrm{air}})$; 1.02 μm for $\theta_{\mathrm{air}} = 33.6°$. This value is in agreement with the experimental value of 1.1 μm. The period was varied with the change in the interference angle θ_{air}. Images in Figs. 3(a)–3(c) show structures fabricated at different interference angles of (a) 10.8°, (b) 21.9°, and (c) 33.6°. As seen, the

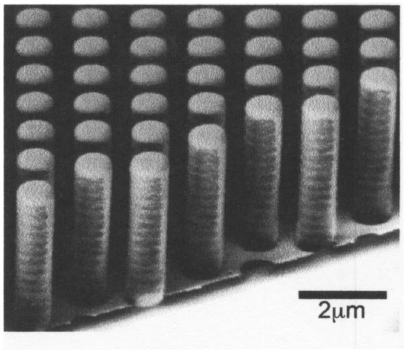

FIG. 4. Close-up view of rods in the structure fabricated by four-beam interference of fs pulses with an interference angle of 21.9°.

period decreases with increasing interference angle, and is in agreement with the value predicted by the calculation. This proves that the structures were fabricated by laser interference.

Figure 3(d) shows the structure fabricated by three-beam interference, in which one of four beams was removed, with an interference angle of 33.6°. In this case a square lattice with the same period as the previous case was again obtained, but the rod cross section is elliptically shaped. This is in agreement with the calculated results. This indicates the possibility of controlling the shape of periodic features (or the ‘‘atomic’’ basis of the lattice) without changing the lattice.

Figure 4 provides a close-up view of the rods fabricated with an interference angle at 21.9°. To obtain this image, the coverglass containing the fabricated structures was deliberately broken and a small fragment was observed. As seen, the rods are slightly bellows-shaped. Distinct ring-like features repeat periodically along each rod with a period about 0.3 μm. This is most likely the result of interference between the incident and reflected beams at the resist–coverglass interface.

In conclusion, we have fabricated 2D periodic structures by multibeam interference of fs pulses. The fabricated rods arranged into a square lattice have high quality and high aspect ratios. We have also demonstrated the possibility of controlling lattice period, as well as rod thickness and shape. The multiphoton nature of the fabrication process was confirmed. Although only relatively simple three- and four-beam interferences were carried out in the present experiments, more beams can be easily added, which allows more complex periodic structures.

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