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Femtosecond laser interference technique with diffractive beam splitter for fabrication of three-dimensional photonic crystals

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A simple optical interference method to fabricate microperiodic structures was demonstrated. Femtosecond laser pulse was split by a diffractive beam splitter and overlapped with two lenses. Temporal overlap of the split femtosecond pulses, which requires 10 μm order accuracy in optical path lengths, was automatically achieved by this optical setup. One-, two-, and three-dimensional periodic microstructures with micrometer-order periods were fabricated using this method. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1391232]

A photonic crystal (PhC) is an artificial structure whose refractive index is periodically modulated. A PhC can alter the propagation property of light with a wavelength of roughly twice that of its period.[1] Due to its promising applications such as integrated optical circuits and thresholdless lasers, PhCs have been intensively investigated.[2,3]

The fabrication of PhCs which work in the visible or near-infrared range is still a challenging topic. The principal method involves semiconductor fabrication technology, which includes lithography, layering, and etching processes. Several sophisticated methods have been developed,[4,5] but they require expensive and large-scale equipment. Other techniques have also been investigated such as laser microfabrication[6,7] and self-organization of microspheres.[8–10] The laser interference technique is also applicable for the fabrication of PhCs, because it produces a periodically modulated optical intensity with a period in the order of its wavelength. The interference of two beams creates a one-dimensional (1D) periodic pattern. By increasing the number of beams, in principle, two-dimensional (2D) and three-dimensional (3D) periodic patterns can be designed. Burns *et al.* have demonstrated an interference technique for arranging small particles into 2D periodic patterns.[11] If periodically modulated optical intensity is transferred to a photoreactive material, a PhC is obtained. Campbell *et al.* have made fcc-type 3D PhC structures in photoresist by overlapping four nanosecond pulses.[12] Shoji *et al.* used five cw beams, which resulted in a hexagonal structure of photopolymerizable resin.[13] These are remarkable accomplishments; however, a complicated optical setup is required for the interference of multiple laser beams, and its precise adjustment is difficult.

In this letter we demonstrate the fabrication of PhCs using multibeam laser interference with a simple optical setup based on a diffractive beam splitter (DBS). The setup is easily applicable for the interference of femtosecond pulses. The images of multidimensional periodic structures fabricated by this method are presented.

The optical setup is schematically shown in Fig. 1. A DBS, which is the key element of this setup, splits the introduced laser beam into several. The split beams are made parallel by a lens, L1, then selected by an aperture array to obtain an aimed interference pattern. The selected beams are gathered by a lens, L2, and create interference at the focused region, where the sample (photoreactive material) is placed. The distance between L1 and L2 was adjusted to make each beam plane wave after the L2.

The DBS used was G1029A (MEMS Optical, Inc.). It splits the input beam into nine beams; in this study only five of them (α, β, γ, δ, and ϵ) were used, as shown in Fig. 1. The combinations of the beams used in the present study are summarized in Table I. The angle θ_{air} is determined by the DBS, L1, and L2. An achromatic lens (Edmond J32883, $f = 175$ mm) was used as L1 and an objective lens (Olympus Uapo/340) was used as L2; so that the θ_{air} was about 24.7°. As seen in the figure, the angle changes to θ_s in the sample according to Snell's law. The intensity of the central beam, I_ϵ, was about 20 times larger than that of the other beams. Since the ideal value of this ratio is 4 for the beam set B3, a homemade filter was used which reduce only the intensity of I_ϵ.

The second harmonic of femtosecond pulses (380 nm, 80 fs, 82 MHz) was used as an irradiation source for fabrication. Irradiation power and duration were adjusted to obtain clear periodic structures: typically about 100 μW (sum of all irradiated beams) and 20 s, respectively. No adjustment for the pulses' temporal overlap was performed. The sample

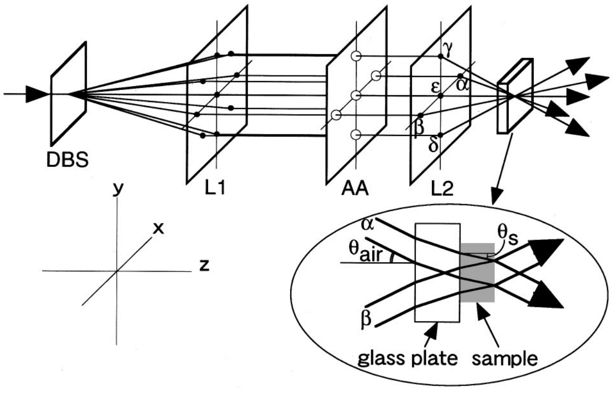

FIG. 1. Optical setup. DBS: diffractive beam splitter, L1 and L2: lenses, and AA: aperture array. The inset shows beam configuration around the sample.

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TABLE I. Selection of beams.

Name	Selected beams
B1	α, β
B2	α, β, γ, δ
B3	α, β, γ, δ, ϵ

material used to make PhCs is a negative photoresist SU-8 (Microlithography Chemical Corp.). It was spin coated on a glass plate forming about a 25-μm-thick film, and prebaked before irradiation. After irradiation, it was postbaked and developed. Periodic structures were eventually obtained.

For observation of the fabricated structures, an optical microscope (Olympus IX-70) and a scanning confocal microscope (Zeiss LSM-410) were used. The confocal microscope was adopted to reveal the 3D structures inside the fabricated region. For that case, a small amount of dye (Rhodamine 6G) was added into the photoresist before exposure and luminescence from the dye contaminated in the solidified resin was detected.

By the beam set B1, the interference of two beams, a grating-like 1D periodic structure was obtained (not shown). Theoretically, the period corresponds to the difference in wave vectors of the two beams, i.e., $\lambda_s/2\sin\theta_s = \lambda_{\mathrm{air}}/2\sin\theta_{\mathrm{air}}$ where λ_s and λ_{air} are the wavelength of the laser in air and in the sample. The experimentally obtained period was about 500 nm. This value is in agreement with the calculated value of 460 nm. This agreement indicates the periodic structure was in fact fabricated by the interference of the two laser beams.

In the case of four beams, the beam set B2, a 2D periodic structure is expected. The image of the obtained structure (top view) is shown in Fig. 2(a). As seen, a four-fold symmetric structure was fabricated. The period was about 700 nm. This is in agreement with the calculated value of $\lambda_s/(\sqrt{2}\sin\theta_s) = 640$ nm. Figure 2(b) shows the cross section of the structure. It should be noted that if the exposure dose was not sufficient, a part of the resin was solidified, but the whole structure was not self-supporting enough to withstand the development procedure.

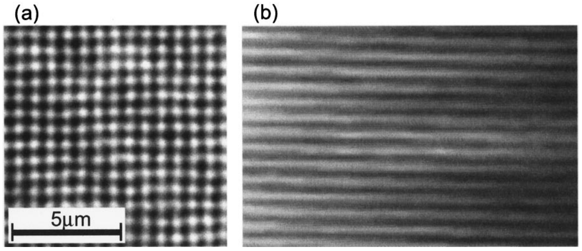

FIG. 2. Image of the structure fabricated by the beam set B2, (a): *xy* cross section and (b): *xz* cross section.

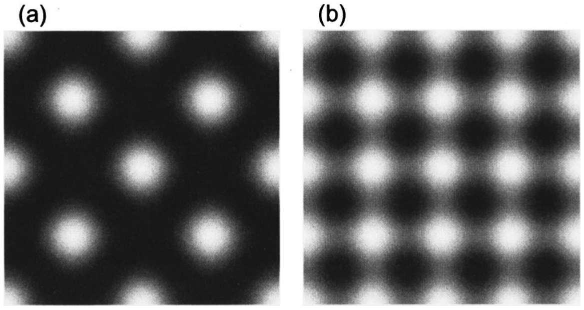

FIG. 3. Calculated intensity distribution by the interference of four beams with different relative phases, (a) all the beams have same phase, and (b) phase of one beam is shifted by π.

The intensity profile of four beam interference I is calculated as

$$I = \langle |E_\alpha + E_\beta + E_\gamma + E_\delta|^2 \rangle, \tag{1}$$

where

$$E_\alpha = E_0 \cos(k\cos\theta z - k\sin\theta x - \omega t + \phi_\alpha),$$

$$E_\beta = E_0 \cos(k\cos\theta z + k\sin\theta x - \omega t + \phi_\beta),$$

$$E_\gamma = E_0 \cos(k\cos\theta z - k\sin\theta y - \omega t + \phi_\gamma),$$

$$E_\delta = E_0 \cos(k\cos\theta z + k\sin\theta y - \omega t + \phi_\delta),$$

where k and ω are the wave vector and angular frequency of the beam, respectively, E_0 is the constant of electric field strength, and ϕ_α, ϕ_β, ϕ_γ, and ϕ_δ are the phases of the beams.[14] The intensity profile I does not depend on z. In the simple case of $\phi_i = 0$ $(i = \alpha, \beta, \gamma, \delta)$, the intensity profile is

$$I = 2|E_0|^2[\cos(k\sin\theta_s x) + \cos(k\sin\theta_s y)\}^2, \tag{2}$$

which is shown in Fig. 3(a). It is four-fold symmetric, and the distance between adjacent peaks is $\lambda_s/(\sqrt{2}\sin\theta_s)$. This intensity profile varies with ϕ. For example, in the case of $\phi_\alpha = \phi_\beta = \phi_\gamma = 0$ and $\phi_\delta = \pi$:

$$I = 2|E_0|^2[\cos^2(k\sin\theta_s x) + \cos^2(k\sin\theta_s y)]. \tag{3}$$

This calculated intensity profile is shown in Fig. 3(b). As seen, it is also four-fold symmetric but its number of peaks is double and the distance between adjacent peaks are $\lambda_s/2\sin\theta_s$, which is $1/\sqrt{2}$ of (a). In our experiments, however, structures with a period about $\lambda_s/(\sqrt{2}\sin\theta_s)$ [Fig. 3(a)] were obtained exclusively. This is explained as follows. In general, the intensity I [Eq. (1)] can be expressed as

$$I = 2|E_0|^2\left[\cos^2\left(Ax + \frac{\alpha-\beta}{2}\right) + \cos^2\left(Ay + \frac{\gamma-\delta}{2}\right) + 2B\cos\left(Ax + \frac{\alpha-\beta}{2}\right)\cos\left(Ay + \frac{\gamma-\delta}{2}\right)\right], \tag{4}$$

where $A = k\sin\theta_s$ and $B = \cos[(\alpha+\beta-(\gamma+\delta))/2]$. Ignoring the position of the peaks, the profile is determined only by the factor B. The intensity of the two type peaks [peaks P1: appear in (a), and peaks P2: appear only in (b)] is $1+B$ and $1-B$, respectively. Only in the case when B is strictly equal to zero, the intensities at the P1 and P2 are identical so that structure similar to Fig. 3(b) is fabricated. In the most cases, B is not equal to zero, thus the intensities at P1 and P2 are different. The less distinct peaks tend to disappear through the development process. Accordingly, structures similar to (a) were obtained exclusively. Further control of optical phases would allow the creation of a structure similar to that shown in (b).

For both beam sets B1 and B2 (Table I), the structures were basically independent of the depth; that is, the same structure was observed in any *xy* plane. This is because the used beams (α, β, γ, δ) have the same wave vector in the *z*

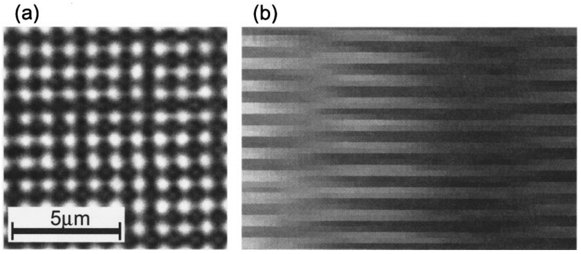

FIG. 4. Image of the structure fabricated by the beam set B3, (a) *xy* cross section and (b) *xz* cross section.

axis. However, the central beam ϵ has a different wave vector in the z axis. As a result, in the case of B3 (α, β, γ, δ, and ϵ) the obtained structure was periodic in the z axis as well as in the x and y axes. Figure 4 shows the structures fabricated by the beam set B3, in which the beam ϵ was added to B2. Figure 4(a) is an xy cross section. It is also four-fold symmetric, but in the middle of the four peaks some solidified objects are seen in the underlayer. This suggests that a 3D periodic pattern was fabricated. The periodicity in z is more clearly seen in Fig. 4(b), which is a zx cross section of the structure.

The advantage of the interference method is its ability to create a periodic structure of large volume through an irradiation process, as well as the uniformity of period. Using a laser microfabrication technique, Sun *et al.*[6] and Cumpston *et al.*[7] have reported the fabrication of 3D PhC structures. In those cases, the structures were fabricated step by step through precisely scanning the sample, therefore requiring much longer time. In addition, the optical setup of the present method is quite simple (the number of optical elements is small), as a result it is stable and easy to adjust. Furthermore, the present method is suitable for the interference of femtosecond pulses. Usually precise adjustments of optical delay are required for each optical path to obtain the temporal overlap of femtosecond pulses, but in the present setup temporal overlap was achieved without any adjustment. A femtosecond laser is preferable for two-photon-absorption fabrication, which is a powerful technique to make micro-3D structures.[15,16] The present method could be applied to two-photon interference fabrication.

In conclusion, we have demonstrated a laser interference method to make 1D, 2D, and 3D periodic structures. This method has advantages of simple optical setup and flexibility in structure through the choice of interfering beams.

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