Diamond photonic band gap synthesis by umbrella holographic lithography

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(Received 20 December 2005; accepted 25 July 2006; published online 7 September 2006)

The authors demonstrate that optical interference lithography yields diamond photonic band gap (PBG) architectures with PBGs as large as 25% when the exposed photoresist is replicated with silicon. This process utilizes five linearly polarized beams propagating from the same half-space (umbrella configuration), a setup considerably simpler than the widely studied counterpropagating four-beam setup. Using the umbrella configuration, this diamond structure is also achieved by two or more exposures using fewer interfering laser beams. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2347112]

Holographic lithography of photonic crystals (PCs) is a fast and inexpensive fabrication process based on the exposure of a photoresist material with an optical field created by the interference of monochromatic laser beams.[1–6] Following an exposure for a time interval $\delta\tau$, the photoresist becomes a mixture of two separate phases, under- and overexposed, with the boundary between these two regions being determined implicitly by the isosurfaces $T=\delta\tau\times I(\mathbf{r})$, where T is the energy threshold for photopolymerization. After the exposure the photoresist is treated with a developer substance which removes selectively only one of the two phases. The structure obtained after the developing stage can be used as a template for the fabrication of photonic band gap materials using established methods such as silicon double inversion.[7] The mininum number of beams required to create a three dimensional interference pattern is four and there are a number of published reports that study in detail this special case.[8–10] All four-beam configurations impose strict requirements on the amplitudes and polarizations of the beams and the ones capable of producing structures with a large photonic band gap (PBG) require a symmetrical distribution of the beams over the 4π solid angle. In this letter we report on a specific five-beam configuration which falls in the so-called "umbrella setup" (see Fig. 1). The main advantage offered by this configuration is the fact that all beam sources are located in the same half-space, thereby allowing the photosensitive material to be mounted on an opaque support. We find simple relations among the phases, amplitudes, and polarizations of the beams such that the generated intensity pattern produces a diamondlike photonic crystal with a full PBG as large as 25% when replicated with silicon.

The field produced by the interference of N monochromatic plane waves of frequency ω, propagation vector $\mathbf{G}_i$, linear polarization vector $\boldsymbol{\epsilon}_i$, phase γ_i, and real amplitude $\mathcal{E}_i$ is given by

$$\mathbf{E}(\mathbf{r},t)=e^{-i\omega t}\sum_{i=0}^{N-1}\mathcal{E}_i\boldsymbol{\epsilon}_i e^{i(\mathbf{G}_i\cdot\mathbf{r}+\gamma_i)}.$$

The generated field intensity is stationary and given by

$$I(\mathbf{r})\equiv\mathbf{E}(\mathbf{r},t)\cdot\mathbf{E}^*(\mathbf{r},t)=\sum_{i=0}^{N-1}\mathcal{E}_i^2+2\sum_{0\leq j<i}^{N-1}\eta_{i,j}\times\cos(\mathbf{K}_{i,j}\cdot\mathbf{r}+\gamma_{i,j}),\quad(1)$$

where $\mathbf{K}_{i,j}\equiv\mathbf{G}_i-\mathbf{G}_j$, $\eta_{i,j}\equiv\mathcal{E}_i\mathcal{E}_j\boldsymbol{\epsilon}_i\cdot\boldsymbol{\epsilon}_j$, and $\gamma_{i,j}\equiv\gamma_i-\gamma_j$. The field intensity can be written as $I(\mathbf{r})=I_0+2\Delta I(\mathbf{r})$, where $I_0=\Sigma_{i=0}^{N-1}\mathcal{E}_i^2$ and

$$\Delta I(\mathbf{r})=\sum_{0\leq j<i}^{N-1}\eta_{i,j}\cos(\mathbf{K}_{i,j}\cdot\mathbf{r}+\gamma_{i,j}).\quad(2)$$

The photonic crystal is a two component structure defined implicitly by the "shape" function $\Theta(I(\mathbf{r})-I_{\rm thr})$, where $I(\mathbf{r})$ is the intensity given above, $I_{\rm thr}$ is a threshold value, and Θ is the Heaviside step function. By convention we assume that the structure of the generated photonic crystal is such

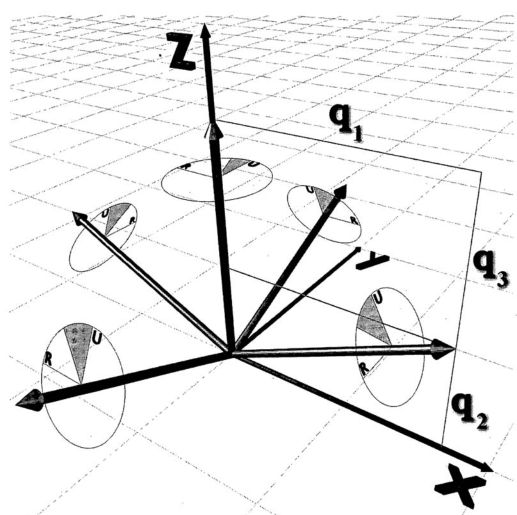

FIG. 1. Umbrella setup: The polarization of each beam is defined with the help of the orthonormal vectors $(\mathbf{U},\mathbf{R})$ and the angle φ made by the polarization vector with $\mathbf{U}$. $\mathbf{U}$, $\mathbf{R}$, and $\mathbf{G}$ are mutually perpendicular and φ—whose corresponding sector is highlighted—is measured in a clockwise direction from $\mathbf{U}$ when $\mathbf{G}$ is the direction of the eyesight. The figure illustrates the beam configuration required by the optimized setup: $\varphi_0\approx 24.3°$, $\varphi_1=\varphi_2=\varphi_3=\varphi_4\approx 41.41°$, and the angle between side and central beams of $\approx 70.53°$.

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0003-6951/2006/89(10)/101117/3/$23.00

that $\varepsilon(\mathbf{r})=1$, where $I(\mathbf{r})<I_{\text{thr}}$ and $\varepsilon(\mathbf{r})>1$, where $I(\mathbf{r})\geq I_{\text{thr}}$.

The linear polarization of each beam is described with the help of an orthonormal pair of vectors $(\mathbf{U},\mathbf{R})$ and an angle φ (see Fig. 1). For a given beam wave vector $\mathbf{G}$ we define two orthonormal vectors, $\mathbf{U}$ ("up") and $\mathbf{R}$ ("right"), such that $\mathbf{R}=\mathbf{G}\times\hat{\mathbf{n}}/|\mathbf{G}\times\hat{\mathbf{n}}|$ and $\mathbf{U}=\mathbf{R}\times\mathbf{G}/|\mathbf{G}|$, where $\hat{\mathbf{n}}=\hat{\mathbf{z}}$ unless $\mathbf{G}||\hat{\mathbf{z}}$ when $\hat{\mathbf{n}}=\hat{\mathbf{y}}$. The polarization vector is expressed in terms of $\mathbf{U}$, $\mathbf{R}$, and φ as $\boldsymbol{\epsilon}=\cos(\varphi)\mathbf{U}+\sin(\varphi)\mathbf{R}$.

In this letter we consider the experimentally suitable five-beam umbrella configuration illustrated in Fig. 1 with incident beam wave vectors: $\mathbf{G}_0=(0,0,q_2+q_3)$, $\mathbf{G}_1=(q_1,0,q_2)$, $\mathbf{G}_2=(-q_1,0,q_2)$, $\mathbf{G}_3=(0,q_1,q_2)$, and $\mathbf{G}_4=(0,-q_1,q_2)$, where q_1, q_2, and q_3 are free parameters to be fixed by the requirements of PBG optimization. The monochromaticity constraint $q_1^2+q_2^2=(q_2+q_3)^2$ implies that only two of the three q_i parameters are independent and $\Delta I(\mathbf{r})$ becomes

$$\eta_{1,0}\cos(q_1x-q_3z+\gamma_{1,0})+\eta_{2,0}\cos(q_1x+q_3z-\gamma_{2,0})+\eta_{3,0}\cos(q_1y-q_3z+\gamma_{3,0})+\eta_{4,0}\cos(q_1y+q_3z-\gamma_{4,0})$$
$$+\eta_{2,1}\cos(2q_1x-\gamma_{2,1})+\eta_{4,3}\cos(2q_1y-\gamma_{4,3})+\eta_{4,2}\cos(q_1(x-y)+\gamma_{4,2})+\eta_{3,1}\cos(q_1(x-y)-\gamma_{3,1})$$
$$+\eta_{3,2}\cos(q_1(x+y)+\gamma_{3,2})+\eta_{4,1}\cos(q_1(x+y)-\gamma_{4,1}).$$

Only four of the phases $\gamma_{i,j}$ that enter $\Delta I(\mathbf{r})$ are independent and we choose them to be $\gamma_{i,0}$ with $i=1.4$. These independent phases cannot be completely eliminated by a spatial translation[11] but we could find a translation that produces $\gamma_{1,0}=\gamma_{2,0}=\gamma$ and $\gamma_{3,0}=\gamma_{4,0}=0$, where γ is the single remaining adjustable phase parameter.

The intensity pattern described by $\Delta I(\mathbf{r})$ is characterized by a long range periodicity with lattice constants $(2\pi/q_1,2\pi/q_1,2\pi/q_3)$ along the (x,y,z) directions, respectively. The precise shape of this tetragonal lattice can be controlled by adjusting the ratio q_1/q_3. A tetragonal lattice characterized by a ratio $c:1$ between the z and x, y lattice constants requires $q_1=q$, $q_2=q/2(c-1/c)$, and $q_3=q/c$. In this case we have $|\mathbf{G}_i|=q/2(c+1/c)$. We further reduce the number of free parameters in $\Delta I(\mathbf{r})$ by assuming that the amplitudes and polarizations of beams 1 to 4 are identical: $\mathcal{E}_1=\mathcal{E}_2=\mathcal{E}_3=\mathcal{E}_4$ and $\varphi_1=\varphi_2=\varphi_3=\varphi_4$. With these simplifying assumptions the intensity pattern depends only on $\mathcal{E}_0$, φ_0, $\mathcal{E}_1$, φ_1, and γ, and the following equalities are true: $\alpha_1\equiv\eta_{1,0}=-\eta_{2,0}$, $\alpha_2\equiv\eta_{3,0}=-\eta_{4,0}$, $\alpha_3\equiv\eta_{2,1}=\eta_{4,3}$, and $\alpha_4\equiv\eta_{3,1}=\eta_{3,2}=\eta_{4,1}=\eta_{4,2}$. With the notations above, $\Delta I(\mathbf{r})$ becomes

$$2\alpha_1\sin(qx)\sin\left(\frac{q}{c}z-\gamma\right)+2\alpha_2\sin(qy)\sin\left(\frac{q}{c}z\right)$$
$$+\alpha_3(\cos(2qx)+\cos(2qy))+2\alpha_4\cos(q(x-y))\cos(\gamma)$$
$$+2\alpha_4\cos(q(x+y))\cos(\gamma). \tag{3}$$

In an earlier analysis[8] we used symmetry considerations to optimize a four-beam intensity pattern for PBG formation. With the notations used in Eq. (3), it can be shown that the optimized diamondlike intensity pattern described in Ref. 8 is proportional to $\Delta I_{\text{diamond}}(\mathbf{r})=\sin(qx+\xi_x)\sin(qz/\sqrt{2})+\cos(qy+\xi_y)\cos(qz/\sqrt{2})$, where q, ξ_x, and ξ_y are arbitrary parameters. This expression can be obtained from Eq. (7) of Ref. 8 by applying a $\pi/4$ rotation around the z axis followed by an arbitrary scale (parameter q) and translation (parameters ξ_x and ξ_y). The intensity pattern of our five-beam umbrella setup, Eq. (3), reduces to $\Delta I_{\text{diamond}}(\mathbf{r})$ if $c=\sqrt{2}$, $\alpha_3=0$, $|\alpha_1|=|\alpha_2|$, and $\gamma=\pi/2$. This choice of c determines the angle between the side and central beams to be $\arctan(2\sqrt{2})\approx70.53^{\circ}$. The second constraint, $\alpha_3=0$, determines the polarization angle φ_1 of the side beams to be $\varphi_1=\pm\frac{1}{2}\arccos\left(\frac{1}{8}\right)=\pm41.4096^{\circ}$. From condition $|\alpha_1|=|\alpha_2|$, we find that the corresponding polarization angle of the central beam is $\varphi_0=\pm\beta_0+m\times90^{\circ}$, where $\beta_0=2\arctan((\sqrt{7}-2)/3)=24.2952^{\circ}$, and m is an integer. For all possible combinations $\{\varphi_0,\varphi_1\}$ we have $|\alpha_1|=|\alpha_2|=\mathcal{E}_0\mathcal{E}_1/2$.

We note that the phases used in the derivations above produce the same intensity pattern (modulo a shift of the origin) as the more general phase distribution, $\gamma_{1,0}=q_1\delta_x-q_3\delta_z+\gamma$, $\gamma_{2,0}=-q_1\delta_x-q_3\delta_z+\gamma$, $\gamma_{3,0}=q_1\delta_y-q_3\delta_z$, and $\gamma_{4,0}=-q_1\delta_y-q_3\delta_z$, where $(\delta_x, \delta_y, \delta_z)$ corresponds to an arbitrary translation. The choice of the best set of phases is determined by the specific experimental setup. A particular example of an equivalent distribution of phases is $\gamma_{1,0}=2\gamma$ and $\gamma_{2,0}=\gamma_{3,0}=\gamma_{4,0}=0$. More specifically, the diamondlike intensity pattern can be obtained by having a single side beam shifted in phase by π relative to the others, or having two counterpropagating side beams shifted by $\pi/2$ relative to the others.

Defining $\tilde{q}\equiv q/\sqrt{2}$, the diamondlike intensity pattern, generated by the umbrella setup using the above constraints, is

$$\Delta I(\mathbf{r})=\mathcal{E}_0\mathcal{E}_1(\sin(qx)\cos(\tilde{q}z)+\sin(qy)\sin(\tilde{q}z)). \tag{4}$$

Clearly the spatial distribution of this intensity pattern is not affected by the ratio $\mathcal{E}_0/\mathcal{E}_1$. This feature offers a significant practical advantage and enables another important design optimization. Indeed, once the phases and polarizations of the beams have been fixed it is useful to adjust the central beam amplitude $\mathcal{E}_0$ to yield the largest contrast in the overall interference pattern. From Eq. (1) this intensity contrast is given by $C\equiv2\max(\Delta I(\mathbf{r}))/I_0\propto\mathcal{E}_0\mathcal{E}_1/(\mathcal{E}_0^2+4\mathcal{E}_1^2)$. This is maximized by choosing $\mathcal{E}_0/\mathcal{E}_1=2$. In the case of the intensity pattern given by Eq. (4), the maximum intensity contrast $C=1/\sqrt{2}\approx71\%$. When all five beams have equal amplitudes, $C=56\%$. An optical field with a large intensity contrast has the advantage that the generated template is less sensitive to imperfections in the photoresist. Based on the values displayed in Table I of Ref. 10 we conclude that our five-beam setup compares very well with the best four-beam fcc configuration which involves elliptically polarized beams.

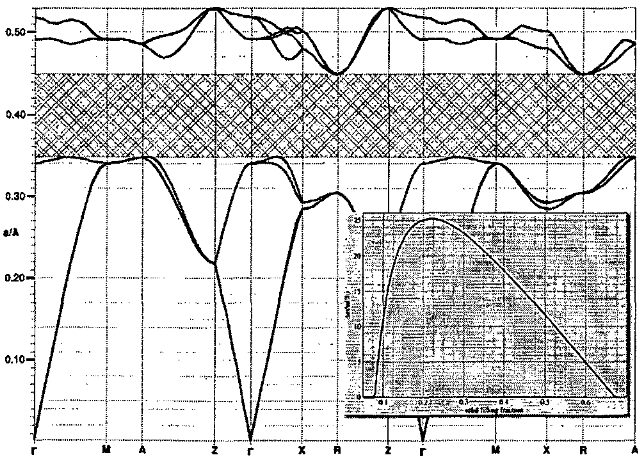

FIG. 2. Band structure of the PC obtained after the exposure of the optimized pattern, Eq. (4), with a scaled threshold, defined as $I_{\rm thr}=\mathcal{E}_0^2+4\mathcal{E}_1^2+2\mathcal{E}_0\mathcal{E}_1 I^s_{\rm thr}$, of $I^s_{\rm thr}=0.68$. The high intensity region has a dielectric constant of $\epsilon=11.9$ and a filling fraction of $\approx 22\%$. The low intensity region has $\epsilon=1$. The full PBG has a relative width of 25.2% and is centered at $a/\lambda \approx 0.4$ where a is the tetragonal lattice constant in the xy plane. The inset shows the variation of the PBG size with the volume fraction of the solid component. The gap remains open for solid filling fractions from 8% to 67% while the scaled threshold varies from $I^s_{\rm thr}\approx 1$ to $I^s_{\rm thr}\approx -0.41$. The gap is greater than 20% for filling fractions from 13% to 36%.

Figure 2 shows the band structure of the photonic crystal made from Si in air with a Si volume fraction of $\approx 22\%$. The inset of Fig. 2 displays the dependence of the size of the full photonic band gap to the volume filling fraction of the solid material. The dependence of the maximum relative size of the photonic band gap on the index of refraction contrast between the two materials is given by the solid curve in Fig. 19 of Ref. 10. TiO_2($n=2.3$) replication would yield a PBG of roughly 8% of the center frequency whereas the PBG vanishes for this diamond architecture as the index contrast approaches 2.0.

Finally, we note that the parameter constraints leading to Eq. (4) correspond physically to a complete decoupling between the side beams. In particular, $\alpha_3=0$ and $\cos(\gamma)=0$ imply that all the terms in Eq. (1), proportional to $\eta_{i,j}$ with $i,j>0$, do not contribute to the optimized optical intensity pattern in Eq. (4). The only important interference is between the central beam and each of the side beams. Consequently, the diamondlike intensity pattern in Eq. (4) can also be obtained using (A) two successive three-beam exposures[12] or (B) four successive two-beam exposures. In case (A) there are two options: (i) Set the phase of all side beams relative to the central beam to be the same; block all but beams 0, 1, and 2 in the first exposure and then, after a 90° phase shift[13] on the central beam, block all but beams 0, 3, and 4 in a second exposure. (ii) Alternatively, use only beams 0, 1, and 2, perform the first exposure, then rotate the sample by 90° about $\hat{z}$ and add the 90° phase shift[13] on the central beam for the second exposure. The modulation pattern created in the photo-resist by these two successive exposures should be identical to the one created by the single five-beam exposure of equal duration (only the background intensity I_0 is different in the two cases). In case (B), the same modulation pattern can be created by four successive exposures with only two beams. In this case the constraint $\alpha_3=0$ is not required; therefore the two beams can be linearly polarized in the same direction. The four-exposure process can be implemented as above, either by blocking all but the central and one side beam during each of the four exposures (in the full umbrella setup) or by using only beams 0 and 1 with three consecutive 90° rotations of the sample between each exposure. Between each exposure, a 90° phase shift[13] is added to the central beam.

In summary, we have demonstrated a five-beam "umbrella" configuration for synthesis of a diamondlike photonic crystal, which, when replicated with silicon,[7] exhibits a 25% full PBG. This umbrella configuration requires only linearly polarized light, but involves a relative phase control between the central beam and side beams when only a single exposure with all beams is performed. To circumvent the complications of relative phase control between different beam directions we have introduced two multiple-exposure protocols that involve only 90° phase shifts of the central beam. This in turn can be achieved with a quarter wave plate or the placement of the sample on a translation stage. Our recipe provides a major experimental simplification of the holographic lithography method for achieving the "holy grail" diamond PBG architecture.

[1] M. Campbell, D. N. Sharp, M. T. Harrison, R. G. Denning, and A. J. Turberfield, Nature (London) **404**, 53 (2000).
[2] Satoru Shoji and Satoshi Kawata, Appl. Phys. Lett. **76**, 2668 (2000).
[3] Yu. V. Miklyaev, D. C. Meisel, A. Blanco, G. Von Freymann, K. Busch, W. Koch, C. Enkrich, M. Deubel, and M. Wegener, Appl. Phys. Lett. **82**, 1284 (2003).
[4] Ivan Divliansky, Theresa S. Mayer, Kito S. Holliday, and Vincent H. Crespi, Appl. Phys. Lett. **82**, 1667 (2003).
[5] X. Wang, J. F. Xu, H. M. Su, Z. H. Zeng, Y. L. Chen, H. Z. Wang, Y. K. Pang, and W. Y. Tam, Appl. Phys. Lett. **82**, 2212 (2003).
[6] D. C. Meisel, M. Wegener, and K. Busch, Phys. Rev. B **70**, 165104 (2004).
[7] N. Tétrault, G. von Freymann, M. Deubel, M. Hermatschweiler, F. Pérez-Willard, S. John, M. Wegener, and G. A. Ozin, Adv. Mater. (Weinheim, Ger.) **18**, 457 (2005).
[8] Ovidiu Toader, Timothy Y. M. Chan, and Sajeev John, Phys. Rev. Lett. **92**, 043905 (2004).
[9] Chaitanya K. Ullal, Martin Maldovan, Edwin L. Thomas, Gang Chen, Yong-Jin Han, and Shu Yang, Appl. Phys. Lett. **84**, 5434 (2004).
[10] Timothy Y. M. Chan, Ovidiu Toader, and Sajeev John, Phys. Rev. E **71**, 046605 (2005).
[11] In the four-beam configuration these phases can always be eliminated by a spatial translation (Ref. 8).
[12] In this case the polarizations of the two counterpropagating side beams can be chosen to be different from the ones demanded by the full five-beam setup.
[13] The 90° phase shift can be achieved with the help of a quarter wave plate rotating from the slow to the fast axis or by translating the sample in the z direction by $3\lambda_0/8$ ($\lambda_0\equiv$ vacuum wavelength of beams).