Fabrication of three-dimensional periodic microstructures in photoresist SU-8 by phase-controlled holographic lithography

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New Journal of Physics 8 (2006) 250
Received 16 August 2006
Published 24 October 2006
Online at http://www.njp.org/
doi:10.1088/1367-2630/8/10/250

Abstract. Three-dimensional (3D) periodic dielectric structures were recorded in a commercial photoresist SU-8 by exposure to intensity patterns created by five interfering coherent femtosecond Ti:sapphire laser pulses. Periodic symmetry of these patterns was controlled by the choice of mutual convergence angles of the beams as well as their phases. The fabricated structures are mechanically and chemically stable, well-connected frameworks of SU-8 regions in air; the smallest dimensions of SU-8 features are in the sub-micrometre range. The structures exhibit a high degree of long-range order across areas as large as 1 mm in cross-section.
1. Introduction

Photonic crystals (PhCs) are periodic dielectric structures comprised of ‘atomic’ dielectric features which facilitate periodic refractive index variation in space and the corresponding modification of dispersion relations for propagating electromagnetic waves [1, 2]. The most notable modifications are photonic band gaps (PBGs) and photonic stop gaps (PSGs), which are spectral ranges forbidden for electromagnetic radiation propagating along any direction, or along selected direction(s), respectively. Engineering of PBG and PSG materials is often a difficult task, especially in the case of three-dimensional (3D) structures intended for application at infrared to visible spectral ranges, because this requires one to assemble dielectric ‘atomic’ features having sub-wavelength (i.e., sub-micrometric) size into ordered 3D arrays spanning many periods along the three directions in space [3, 4]. Despite the fact that these difficulties have prompted successful development of simpler 1D and 2D PhC structures, it must be noted that some functionality of PhCs (for example, optical microcavities or waveguides in air is only achievable with 3D structures [5]). Fabrication technique based on optical holographic lithography [6]–[8] is among the most practical and elegant approaches to the fabrication of extended periodic 3D micro- and nano-structures. Although it can mostly be applied to low-refractive index materials, such as photoresists and photopolymerizing liquid resins, it allows us to obtain structures with symmetries corresponding to all fourteen 3D Bravais lattices [9]. Moreover, optical fields with symmetries resembling those of 3D quasi-crystals, e.g., Penrose patterns [10] can be generated and recorded as well. The recording essentially involves a single-step exposure to a spatially periodic optical field created by multiple interfering laser beams or pulses. Depending on the laser wavelength, the exposure can be achieved via linear or nonlinear absorption. This versatility makes holographic lithography a highly attractive method for future mass-production of periodic micro-structures. The low index contrast of laser-microfabricated structures has been tackled recently by double infiltration with silicon (refractive index \( n = 3.5 \) at infrared wavelengths) which produced structures possessing full PBG [11]. This approach is obviously also suitable to holographically fabricated low-index templates, whose symmetries and parameters define those of the final structures, and therefore template tailoring is the important first stage of the fabrication process.
Various optical schemes used in implementing the holographic lithography usually differ in the ways the multiple interfering beams are obtained and converged on the sample. The most straightforward approach, which uses multiple beamsplitters for obtaining a set of coherent beamlets, delay lines to ensure their temporal matching, as well as multiple mirrors and a focusing lens to ensure their spatial overlap, is also the most cumbersome to align and highly susceptible to mechanical disturbances. Recently, a much simpler scheme, based on a phase mask was successfully applied [12, 13]. The phase mask is a periodic dielectric structure attached to the surface of the fabricated sample, which splits a single incident beam into several spatially and temporally overlapping waves propagating along different directions, and creating a periodic 3D intensity pattern in the sample. Benefits of this scheme are its simplicity, insensitivity to mechanical disturbances, and the possibility to use non-laser light (e.g., spectrally filtered lamp radiation). However, this scheme can produce only one kind of lattice symmetry with the given phase mask, and it relies on van der Waals bonding between the phase mask and the sample (e.g., thin photoresist film), which therefore must be prepared to have an atomically smooth, defect-free surface. Another similar scheme uses a refracting prism for obtaining and overlapping the multiple beamlets in the sample [14].

Earlier we introduced a scheme of holographic lithography which uses a diffractive optical element (DOE) for obtaining the set of multiple, perfectly synchronized beamlets, from which various beamlets required to create the desired lattice symmetry can be selected using an amplitude mask (a set of apertures), and overlapped on the sample with a lens [15]. This scheme has proven highly stable mechanically, requires no physical contact with the sample, and most importantly, allows the choice of various lattice symmetries by blocking/unblocking of various beamlets in situ. Using this scheme, various photonic periodic structures were successfully fabricated via two-photon absorption (TPA) in a commercially available high-aspect ratio photoresist SU-8 [16]. DOE is essentially a 2D diffraction grating which can be fabricated or obtained commercially. In our studies (including this one) we use cheap, commercially available DOEs, which, although not of the highest quality, allow fabrication of high quality structures. The main limitation of the scheme employing DOE is a relatively low mutual beamlet convergence angle, which in many cases results in strong anisotropy of the lattice period—a short, sub-micrometric period in the transverse plane perpendicular to the common convergence direction of the beamlets, and a longer period, typically of a few micrometres, along that direction. However, this circumstance may prove advantageous for applications, which would require the dominant photonic band functionalities at different spectral regions along different directions—for example a PBG/PSG in the visible spectral range in the transverse plane, and a PBG/PSG at infrared wavelengths along the direction normal to that plane. An additional, and less explored, benefit of the DOE-based holographic lithography is the possibility to adjust phases of the interfering beamlets, which allows in situ modification of the interference patterns, and expands the available range 3D symmetries. In this study, we extend our earlier studies of holographic lithography in SU-8 [17]–[20] by demonstrating the capabilities of phase control for tailoring 3D structure symmetry, using as a test platform the arrangement of five interfering beams that deliver femtosecond laser pulses to the SU-8 sample. The structures fabricated are extended frameworks of well-connected, mechanically stable, and chemically robust SU-8 features with sub-micrometre dimensions in air. Their long-range periodicity was confirmed by scanning electron microscopy (SEM) and optical transmission and reflection measurements at infrared wavelengths, and indicates the possibility to apply these structures as elements of photonic and micro-mechanical systems.
2. Experimental details

2.1. Implementation of holographic lithography using DOE

The optical scheme of holographic lithography used is schematically illustrated in figure 1(a). The beam of a femtosecond Ti : sapphire laser system, which delivers 150 fs pulses centred at the 800 nm wavelength at a repetition rate of 1 KHz, passes through a DOE, where it is split into a set of diverging multiple beamlets by diffraction. The set of beamlets is subsequently collimated by a lens, and passed through the amplitude mask which selects the beamlets required for obtaining the desired 3D structure. Optical path length between the DOE and the sample is typically 20 cm. As the beamlets propagate in free space, they can be optionally passed through variable phase retarder (PR) plates which set their mutual phases, and as will be illustrated below, alters the 3D interference patterns. The parallel, phase-controlled set of beamlets is then focused into the sample using a focusing lens having numerical aperture \( NA = 0.75 \). The use of DOE produces tilted laser pulses whose mutual coherency and zero time delay is maintained across their entire cross-sectional areas despite the different directions they converge on the focal region of the focusing lens. Hence, the interference occurs in the entire area of the focal spot. More details about the five-beam experimental configuration and the set-up used can be found in our earlier study [8, 21].

The DOE used was G1023A (MEMS Optical, Inc.) and is shown in figure 1(b). The symmetry of the pattern seen in the optical micrograph of the DOE makes it obvious that the laser beam transmitted through the DOE will form a diffraction pattern having a square symmetry. Indeed, transmitting a HeNe laser beam through DOE, produced a \( 6 \times 6 \) square matrix of laser spots of nearly equal intensity. A closer inspection of the DOE has also revealed that its overall optical quality is mediocre. For example, it was found that surfaces of the DOE are not parallel, which resulted in phase distortions and bending of the interference fringes which otherwise should be straight, seen by monitoring selected planes of the interference patterns. To compensate for these distortions, PR plates (glass slides with adjustable tilt angles) were used as illustrated in figure 1(a). Similar plates, sputtered by a variable thickness (0–50 nm) gold film, were also used as neutral density filters to set optimum intensities of the interfering beams and maximize the contrast of the interference pattern.

The main factor that can potentially degrade the contrast and long-range uniformity of the interference pattern is the above-mentioned phase distortions in the DOE. Additional distortions may come from geometrical and chromatic aberrations caused by the focusing lens, but in our experiments they played no apparent role.

Figure 1(c) outlines the arrangement of the recording beams. All beams are polarized linearly along the same direction, which can be identified as the direction of line between the points 1–3 (or 2–4) in the figure. The symmetry of the interference pattern is determined by the number of recording beams (four or five) and their convergence conditions, defined by the angle \( \theta \) between the beam direction and the main optical axis of the optical system. The four-beam configuration (left-hand side) employs four beams with the same angle \( \theta \), located on the corners of a square, while the five-beam configuration (right-hand side) has an additional beam at the centre of the square (\( \theta = 0 \)). The symmetric four-beam configuration is not used in the experiments, since it can only produce a 2D intensity pattern. It is included here because it is useful as a reference in the numerical analysis (see the next section). The symmetric five-beam configuration with adjustable phases is capable of delivering a range of 3D interference patterns and was used in the...
experiments. One of the main practical requirements for any interference pattern to be recorded is the maximum fringe contrast. Based on the results of numerical simulations, this parameter was optimized by adjusting the intensity ratio between the central and peripheral beamlets. For scalar focusing conditions when beam depolarization at the focus is negligible, this ratio was set to the theoretically deduced optimum value of 1 : 5.

By mounting a CCD camera in the place of sample (and using a less-powerful focusing lens for the imaging than the lens used for the fabrication), one could monitor 2D intensity patterns at various positions along the $z$-axis, and thus verify the qualitative correspondence between the numerically simulated and actual intensity patterns without the need to actually record and analyse the structures. Occasional distortions found in these patterns were analysed, and their sources identified. Most often these distortions were due to spatial nonuniformities of the phases of the beams occurring in the focusing optics and DOE. This monitoring allowed us to minimize the distortions and deliver nearly distortion-free images [22].
2.2. Photoresist SU-8 samples

The samples fabricated are films of an ultra-thick negative polymeric photoresist SU-8 (Microchem), deposited on glass substrates by spin-coating. The thickness of the SU-8 films varied from sample to sample, but typically was kept up to 25 µm. The samples were mounted perpendicular to the main axis of the optical system (which is parallel to the propagation direction of the initial laser beam and the collimated beamlets), such that 3D interference field in the focal region of the lens overlapped fully the SU-8 film and partially spread into the glass substrate. This condition ensured adhesion of the fabricated structures to the glass substrate. SU-8 is known to be optically transparent at the 800 nm wavelength in the linear regime, which ensured essentially uniform penetration of the interference field into the SU-8 film. At an appropriately chosen laser pulse intensity, a small fraction of the incident radiation is absorbed by the photoinitiator in SU-8 due to a nonlinear process, which in SU-8 at 800 nm wavelength is often ascribed to TPA [16]. The absorption triggers local photochemical reactions leading to polymer cross-linking during the subsequent post-exposure bake step. Polymer cross-linking is also stimulated by optically induced heating which leads to cross-linking of SU-8 even prior to the post-exposure bake [23]. Altogether, the nonlinear optical absorption, heating and post-exposure thermal treatment lead to cross-linking in SU-8 in the regions where local exposure dose exceeds a certain threshold value. The cross-linked regions thus are rendered insoluble during the subsequent wet development, which dissolves and removes only the lesser exposed regions. With appropriately chosen interference pattern and pulse energy/exposure dose, a cross-connected 3D network of solid SU-8 features in air can be obtained attached to the glass substrate. SU-8 is designed for lithographic fabrication of micromechanical systems and is therefore robust mechanically and chemically. Thus, as-fabricated SU-8 provides a stable platform for the fabrication of micromechanical systems using the method described above. Its refractive index \( n \approx 1.6 \) is considerably higher than that of air \( n = 1 \), sufficient for some PhC applications which do not rely on complete PBG.

3. Results and discussion

3.1. Five-beam interference patterns and role of phase control

In general, spatial distribution \( I(\mathbf{r}) \) of the light intensity used for recording periodic structures resembling holograms results from superposition of plane coherent waves and can be expressed as:

\[
I(\mathbf{r}) = \sum_{n,m} E_n e^{-i(k_n \cdot \mathbf{r} + \delta_n)} \cdot E_m^* e^{i(k_m \cdot \mathbf{r} + \delta_m)},
\]

where \( E \) is the optical electric field vector (the star symbol denotes complex conjugate), \( \mathbf{r} \) is the radius vector, \( k \) is the wavevector, and \( n = m \) represents the number of interfering beams. The phases of beams are given by \( \delta_{n,m} = 0 \). For the four and five-beam configurations depicted in figure 1(c), interference patterns simulated by equation (1) are shown in figure 2. The same figure illustrates the effect of phase control imposed on selected beams (see the figure caption for details). In accordance to the experimental conditions, calculations were carried out for beams having identical linear polarizations parallel to the axes 1–3 (or 2–4) in figure 1(c). Hence the
total field had negligible longitudinal component parallel to the main optical axis of the system. Before proceeding further, it is helpful to note that the symmetric four-beam configuration can only produce 2D intensity patterns (whose lattice period along the optical axis of the system can be regarded as infinite). The same is true for symmetric arrangement of any number of beams, e.g., triangular or hexagonal, having equal components of the \( k \)-vector along a certain direction, which in our case is coincident with the main optical axis of the system, and is characterized by infinite lattice period. This condition is violated by adding the central beam to the four beams. Thus, the five-beam configuration produces a 3D periodic lattice with finite period along the main optical axis, which (along any direction) is inversely proportional to the minimum difference between the components of the \( k \)-vectors existing along that direction. For beams converging at moderate angles this difference is small, and as a consequence, the lattice period along the main optical axis becomes much longer than in the plane perpendicular to it.

Equation (1) and figure 2 already give a hint that mutual phases of the beams influence the interference pattern. A visual proof of this prediction is given by the 2D intensity patterns of five beams shown in figure 3. The experimental patterns were taken at different planes using a CCD camera mounted in place of the sample as described in the previous section. The simulated patterns were obtained from equation (1). Figure 3(a) shows five-beam same-phase interference intensity pattern and its selected 2D planes calculated according to equation (1) in comparison with the 2D intensity distribution registered in the similar planes using a CCD as described above. A closer look into these distributions allows one to recognize their body-centred tetragonal (bct) point lattice symmetry. Figure 3(b) shows the analogous result for the case where phases of beams 1 and 3 (see beam designations in figure 1) were shifted by the amount \( \pi/2 \) by tilting the PR plates. Again, good matching between the calculated and experimental patterns is observed.

Figure 2. Simulated interference patterns for four- (a, b) and five-beam (c, d) configurations. Panels (a) and (c): all beams having the same phase, panels (b) and (d): phases of beams 2 and 4 (or 1 and 3) shown in figure 1(c) are shifted by amount \( \pi/2 \).
Figure 3. Comparison between interference patterns of (a) five beams having the same phase, and (b) phases of the two side beams (1, 3 or 2, 4 in figure 1(c)) shifted by $\pi/2$. In both panels, perspective view of the calculated 3D intensity distribution is shown on the left-hand side, its selected 2D $x$–$y$ planes shown in the middle, and experimental images of the corresponding planes are shown on the right-hand side.

Under careful examination the obtained 3D pattern can be identified as having a point lattice whose symmetry can be informally categorized as face-centred tetragonal (fct) and an ‘atomic’ base consisting of two spatially shifted ‘atoms’. A particular case of fct point lattice is the face-centred cubic (fcc) lattice, which has cubic cell symmetry and, if combined with a two-atom basis, forms a so-called diamond structure. The diamond structure is highly desired as an architecture for 3D PhCs due to its widely known tendency towards opening of spectrally wide PBG and PSG [24]. The pattern shown in figure 3(b) can be regarded as a diamond-like structure. The fct symmetry of this pattern results from the above described stretching of the cubic cell along the main axis of the optical system. This elongation will decrease with increasing convergence angle of the peripheral beamlets. However, even in the presence of some elongation and fct symmetry, periodic structures may still display strong PBG properties, as was demonstrated recently for structures composed of spirals, also belong to the class of diamond-like architectures [25]. Therefore the result shown in figure 3(b) indicates the possibility of fast
prototyping of diamond-like periodic structures using the five-beam holographic lithography with phase control.

3.2. Five-beam holographic lithography in SU-8

Figure 4(a) shows SEM images of the 3D periodic structure fabricated using the above-described configuration. During the fabrication SU-8 was exposed to pulses having 24 µJ energy (all five beams combined) for \( t = 90 \) s time interval. The beams’ phases were set uniform, and correspondingly, the structure had bct point lattice symmetry. Its lattice periods in the \( x-y \) plane and along the \( z \)-axis direction are 1.4 and 8.0 µm respectively, which corresponds to the beam convergence half-angle \( \theta = 34^\circ \). The same values of periods were determined at various locations of the exposed region of SU-8 which had diameter of about 1 mm in the \( x-y \) plane. This result illustrates good uniformity and high degree of the long-range order in the sample.

We note that fabrication of diamond-like patterns was less successful; structures resulting after the development and post-processing were most often distorted due to capillary drainage forces despite our best efforts to minimize their effects using methods developed and described earlier [22]. The poor mechanical stability of diamond-like structures most likely comes from their ‘two-atom’ basis formed by two ellipsoidal dielectric features. Non-ellipsoidal (or non-spherical) overall shape of the two-particle composite prevents formation of well-balanced connections with all neighbouring particles. Hence, the structure lacks mechanical equilibrium and can be easier destroyed by factors like capillary forces. In this context, it is interesting to note that in simpler fcc or bcc structures which have only a single ellipsoidal or spherical ‘atom’ at each lattice point, equilibrium in connecting with the neighbouring ‘atoms’ is more reliable. Natural or artificial opals are well-known examples of mechanically stable, extended periodic structures. Artificial opals can even self-organize from chaotic colloidal suspensions of spherical particles.
Figure 5. (a) Schematic layout of the infrared microscope unit attached to the FT-IR spectrometer; L1, L2: Cassegrainian objectives used to irradiate the sample and collect the transmitted/reflected radiation. (b) Thin-lens equivalent of a Cassegrainian objective; $\alpha_{\text{min, max}}$: minimum and maximum acceptance angles describing ring-like aperture of the objective. The blocked central sector of the aperture is shown by the grey circle. The sample oriented normal to the $y$-axis is probed along directions described by the range of acceptance angles; normally incident radiation is absent in the incident beam.

3.3. Characterization of optical properties

We have analysed optical properties of the structure depicted in the previous figure, searching for signatures of photonic bands. Due to the relatively low index contrast, the most likely manifestation of photonic band dispersion in this sample is PSG, which can be identified from spectrally matched regions of optical attenuation and enhanced reflectivity. Transmission and reflection spectra were measured using FT/IR-6000TM-M (Jasco) Fourier-transform infrared (FT-IR) spectrometer equipped with an infrared microscope attachment. The use of microscope enables convenient performance of optical measurements on small (a few micrometres in diameter) samples or in small regions of large samples. Proper interpretation of spectra measured in PBG materials using infrared microscope requires some insight into its basic functioning. Figure 5(a) shows layout of the experimental system. Radiation of a broadband light source is
focused on the sample by a Cassegrainian reflection objectives L1 (in transmission mode) or L2 (in reflection mode). The same Cassegrainian objectives collect the reflected (or transmitted) signal, which is coupled into a FT-IR spectrometer for spectral analysis. From the 2D depiction used in figure 5(a), one can understand that aperture of a Cassegrainian objective is partially blocked at the centre by a circular projection of the back-reflecting spherical mirror. Hence, the probing beam converges from the objective on the sample as a hollow cone. Accordingly, the transmitted/reflected light is collected into the angular acceptance range characterized by the minimum and maximum acceptance angles. The objectives used in our set-up have minimum and maximum acceptance angles of $\alpha_{\text{min}} = 16^\circ$ and $\alpha_{\text{max}} = 32^\circ$, respectively.

A thin lens equivalent of a Cassegrainian objective would have a ring-like aperture determined by the acceptance angles as is illustrated in figure 5(b). In our experiments the most important feature of a Cassegrainian objective is the circumstance that even if the sample is oriented normal to y-axis (figure 5(b)), the normally incident radiation is eliminated in the objective. Instead, the probing radiation has a wide range of incidence directions. Since optical properties of PBG materials are strongly direction-dependent, their measured transmission and reflection spectra should be treated as a superposition of spectra measured along multiple propagation directions imposed by focusing and collecting optics. The superposition (or angular averaging) usually produces spectral shifts, deformations and loss of strength of the spectral features. Therefore, one should expect somewhat degraded optical properties when using micro-FT-IR set-up for the characterization of PBG materials. This factor will be taken into account in the theoretical analysis of our data.

In the sample concerned, the intended direction of optical measurements was along the y-axis (i.e., parallel to either vertical or horizontal directions in the top-left and right panels in figure 4). Longer lattice period in the z-axis direction would result in dominant PSG features at mid-infrared wavelengths (expected around 15–20 $\mu$m), which is outside the technically available spectral range. For measurements in the x–y plane it is essential to couple the probing optical radiation in and out of the sample with minimum losses due to random scattering, which is another factor responsible for degraded quality of the optical spectra. As-fabricated, holographic structures do not have well-defined planar edges, which should therefore be formed by cutting the sample twice to form a narrow stripe having width of about 20–30 $\mu$m, and height corresponding to the thickness of the SU-8 film (about 20 $\mu$m). Despite all efforts, the cut edges of the samples were far from perfect and produced a significant scattering which resulted in diminished strength of the features arising due to the photonic band dispersion.

As illustrated by the measured spectra in figure 6(a), in the wavelength range of 2.5–4.0 $\mu$m a region of higher reflectivity is seen at approximately same spectral position as the region of optical attenuation. Within this region, finer spectral variations in the reflectivity and transmission occur in a clearly anticorrelated manner. This behaviour, and the general spectral matching between optical attenuation and enhanced reflectivity regions are indicative of PSG. In the present experiments, we do not try to estimate magnitudes of the reflectivity peaks and transmission dips in absolute units due to difficulties in acquiring reliable reference spectra (especially for the transmission measurements) needed in order to properly normalize the data. However, our rough estimate is that strength of these features is only a few per cent. Despite their relative weakness, we believe that quality of the structure is masked by the random scattering at the cleaved edges and by the wide range of probing directions used in the measurements.

Figure 6(b) shows the transmission and reflection spectra calculated theoretically using Finite-Difference Time-Domain (FDTD) technique for a model structure with parameters.
Figure 6. (a) Transmission and reflection spectra of the sample measured using FT-IR set-up. The spectra were corrected for the residual linear absorption of SU-8 and spectral variations of the source intensity. (b) Transmission and reflection spectra calculated by FDTD technique for a model structure closely resembling the sample on which measurements were conducted, for normally incident light propagating along the $y$-axis (for more details see figure 7). (c) and (d) Calculations repeated for the same sample in the case of multiple incidence directions approximating the conical beam of a Cassegrainian objective. The grey–yellow–grey regions superimposed on the spectra are to emphasize spectral matching between the features in measured and calculated data.
Figure 7. The model used in the FDTD calculations, the 3D colour map shows unit cell of a dielectric structure generated in a separate calculation based on equation (1), solid parts are filled by solidified SU-8, empty parts are filled with air, in the simplest case the structure is irradiated by a normally incident plane wave; the intensity of the wave propagated through the structure (10 cells) is measured by the detector plane, and is normalized by the incident intensity to deduce the transmission. Another detector plane (not shown) in front of the structure is used to register the reflected intensity and deduce the reflection.

similarity between the calculated pattern and the pattern seen in figure 4. A single cell of the structure thus generated is depicted in figure 7. Its lattice periods match those of the real structure. It can be noticed that air cavities in the calculated structure are isolated from each other by thin walls. In practice such a structure would be not permeable to the SU-8 developer, and hence not feasible to fabricate. However, for the theoretical calculations this structure is a valid model, since its dielectric filling factor (and average refractive index) are only marginally larger than that of the real structure. Total thickness of the sample along the y-axis direction is 10 unit cells. In the experiments, most likely a larger thickness is probed, but since in practice the thickness depends on factors like sample position and orientation, it cannot be determined exactly. For qualitative comparison with experiment, and in order to reduce the time needed for computations, we use relatively thin model sample. Periodicity of the structure is accounted for by imposing periodic (in some cases, Bloch) boundary conditions along the x- and z-axes.

First we model transmission and reflection spectra of the sample in the case of probing by a plane wave incident normally on the sample (along the y-axis). The spectra are shown in figure 6(b) and clearly reveal a PSG region centred near the wavelength of 3.5 μm; the amplitude of the transmission/reflection features reaches about 95% level. The PSG spectral region contains no finer spectral variations. Although the central PSG wavelength predicted
by the calculations is close to that observed experimentally, spectral shapes and amplitudes of the PSG differ strongly. To add more realism to the simulations, we have incorporated optical probing along several directions belonging to the acceptance cone of the Cassegrainian objective. Returning to figure 5, each incidence direction within the acceptance angle can be characterized by a pair of angles \((\alpha, \phi)\). As a crude approximation of the beam focused by a Cassegrainian objective, we have chosen twelve plane waves incident at the same angle \(\alpha_{\text{ave}} = 24^\circ\), i.e., in the middle of the acceptance cone, and with angles \(\phi = n\pi/12, \ n = 0 \ldots 11\), i.e., at every 30\(^\circ\) in the azimuthal degree of freedom. For periodic structure it is sufficient to consider the angular range of \(\phi = 0 \ldots \pi/2\), which comprises four sources and correspondingly, requires performance of four calculations. In each calculation, Bloch boundary conditions were imposed in the \(x-z\) plane according to the incidence angle of the source. Finally superposition of the angularly resolved spectra was found by their averaging.

The spectra simulated using these assumptions are shown in figures 6(c) and (d). Although the central wavelength of the PSG region has remained similar as before, this region has broadened spectrally and acquired some fine structure. The magnitude of transmission and reflection features has decreased considerably to the level of about 50\%. Thus, inclusion of a range of oblique incidence angles and exclusion of the normal incidence angle has led to quite drastic transformation of the simulated spectra. By comparing these spectra to the experimental data shown in figure 6(a), one can notice qualitative similarity between these data. Approximate correspondence between the spectral variations in the measured and calculated spectra is emphasized by shaded rectangles superimposed on the plots. We have verified that noticeable spectral splitting of the PSG transmission dips and reflection peaks occurs essentially due to the superposition of spectra for \(\phi = 0\) (incidence in the \(x-z\) plane) and for \(\phi = \pm \pi/2\) (incidence in the \(y-z\) plane), each of whom produce spectrally distinct dips/peaks, and their average becomes a doublet. By including even more incident waves, in particular multiple incidence angles \(\alpha\), one can possibly improve the theoretical simulations. However, we do not attempt to do this here because of the longer time needed for the simulations, and the fact that experiments are also affected by other circumstances, for example orientation of the sample with respect to the probing beam, which are not easy to assess. At this point, we just stress that our experiments and modelling agree on a qualitative level: calculations assuming normal incidence predict correctly the central wavelength of the PSG, while calculations assuming a range of incidence directions predict correctly the central PSG wavelength and provide reasonable predictions regarding the spectral shapes of transmission/reflection features measured using micro-FT-IR set-up. Differences between the experiment and modelling can be attributed to many factors. The modelling accounts for the angular dependence of the incident radiation, while angular dependence of the signal collection is ignored. On the practical side, radiation losses due to random scattering at the edges of the sample remain the most likely source of the deviations. Formation of smooth edges in the structures fabricated by holographic lithography, either before or after the fabrication, still remains an issue to be addressed.

4. Conclusions

We have fabricated 3D periodic dielectric structures in commercial photoresist SU-8 by the holographic lithography technique which used five interfering femtosecond Ti : sapphire laser pulses with controllable convergence angles, amplitudes and phases. The structures fabricated
are extended frameworks of well-connected, mechanically stable, and chemically robust SU-8 features with sub-micrometre dimensions in air. Their long-range periodicity was confirmed by SEM and optical transmission and reflection measurements at infrared wavelengths, as well as theoretical simulation using FDTD technique. These results indicate the possibility to apply these structures as elements of photonic and micro-mechanical systems.

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