Nanopatterning by Laser Interference Lithography: Applications to Optical Devices

Jung-Hun Seo¹, Jung Ho Park², Seong-II Kim³, Bang Ju Park⁴, Zhenqiang Ma¹,* Jinnil Choi⁵,* and Byeong-Kwon Ju²,*

¹Department of Electrical and Computer Engineering, University of Wisconsin-Madison, Madison, Wisconsin 53706, USA
²Display and Nanosystem Laboratory, School of Engineering, Korea University, Seoul 136-713, Republic of Korea
³Center for Materials Architecturing, Korea Institute of Science and Technology, Seoul 136-791, Republic of Korea
⁴Department of Electronic Engineering, Gachon University, Seongnam 461-701, Republic of Korea
⁵Department of Mechanical Engineering, Hanbat National University, Daejeon 305-719, Republic of Korea

A systematic review, covering fabrication of nanoscale patterns by laser interference lithography (LIL) and their applications for optical devices is provided. LIL is a patterning method. It is a simple, quick process over a large area without using a mask. LIL is a powerful technique for the definition of large-area, nanometer-scale, periodically patterned structures. Patterns are recorded in a light-sensitive medium that responds nonlinearly to the intensity distribution associated with the interference of two or more coherent beams of light. The photoresist patterns produced with LIL are the platform for further fabrication of nanostructures and growth of functional materials used as the building blocks for devices. Demonstration of optical and photonic devices by LIL is reviewed such as directed nanophotonics and surface plasmon resonance (SPR) or large area membrane reflectors and anti-reflectors. Perspective on future directions for LIL and emerging applications in other fields are presented.

Keywords: Laser Interference Lithography, Optical Devices, Reflectors, Anti-Reflectors, Color Filters.

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1. INTRODUCTION

Laser interference lithography (LIL), the maskless exposure of a photore sist layer with two or more coherent light beams, provides a facile, inexpensive, large-area nanolithography technique. Some groups refer to interferometric lithography (IL) interchangeably as interference lithography and holographic lithography.¹⁻⁷ The capabilities and techniques of IL have been reviewed elsewhere⁸⁻¹⁰ and are only briefly covered here. The purpose of this review is to examine various fields in which IL is impacting nanostructure research. IL shares this large-area capability with advanced optical lithography, today’s integrated circuits are at the 45-nm node;¹¹ nanoimprint lithography;¹²,¹³ and various self-assembly approaches such as nanosphere lithography and surface plasmon lithography.¹⁴⁻¹⁷ However, each of these patterning techniques has certain limitations that prevent widespread adoption. The modern lithography tools for generating nanopatterns are so delicate and expensive that they require either highly skilled operators or a large budget for maintaining the system.¹⁸,¹⁹ Nanosphere lithography,¹²¹⁻²⁴ which uses the spaces between monodisperse colloidal spheres deposited or spun on a densely patterned surface, often lacks long-range order, due to the tendency to nucleate uncorrelated domains on the substrate, which yields relatively small (1~100 um) grain boundaries.
Jung-Hun Seo received his B.S. and M.S. degrees in electronics and electrical engineering from Korea University, Seoul, Republic of Korea, in 2006, 2008, respectively. Since 2009, he is working toward a Ph.D. in electrical and computer engineering at the University of Wisconsin-Madison, USA. He's authored and coauthored more than 30 peer-review papers. His current research interests are fast flexible electronics and wide bandgap HBTs.

Jung Ho Park received an M.S. degree in electronics and electrical engineering from Korea University, Seoul, Korea, in 2009. Since 2009, he has been working toward a Ph.D. in electronics and electrical engineering at Korea University, Seoul, Korea. His current research interests include flexible electronics with hybrid thin-film transistors inverter and nano-devices using Laser Interference Lithography (LIL).

Seong-Il Kim received his M.S. degree in physics from Korea Advanced Institute of Science and technology (KAIST) in 1986 and the Ph.D. degree in physics from KAIST in 1994. From 1986, he has been working at Korea Institute of Science and Technology (KIST) as a principal research scientist. Since 2001, he has been a visiting professor in Korea University. His current research interests include solar cell, graphene related devices, FDTD and semiconductor device modeling.

Bang Ju Park received his M.S. degree in electronic engineering and Ph.D. degree in radio engineering from Kyunghee University, Republic of Korea. In 1995, he was a visiting researcher at Harvard University, USA. Currently, he is a professor at the department of Electronic Engineering, Gachon University, Korea. His research interests include cognitive radio technology.

Zhenqiang Ma received a B.S. degree in applied physics and a B.E. degree in electrical engineering from Tsinghua University, Beijing, China, both in 1991. He received an M.S./M.S.E. degree in nuclear science/electrical engineering and a Ph.D. degree in electrical engineering from the University of Michigan, Ann Arbor, in 1997 and 2001, respectively. Since 2002, he has been with the Department of Electrical and Computer Engineering, University of Wisconsin, Madison, as a Professor. He is the author or coauthor of more than 120 peer review papers and book chapters. His current research interests include semiconductor materials, processing and heterogeneous integration, semiconductor device physics and technologies, high-speed electronic and optoelectronic devices, CMOS integrations with lasers and with micro-/nano electromechanical systems (MEMS/NEMS), nanophotonics based on transferable semiconductor membranes, high-speed flextronics, nanoscale semiconductor devices, unconventional multispectral imaging, photovoltaics, isotope betavoltaics, and high frequency RF integrated circuits.
Intensive nanoscience research has progressed with serial writing tools such as electron-beam lithography (EBL),\textsuperscript{25} ion-beam lithography (IBL),\textsuperscript{26–28} and atomic force microscopy (AFM).\textsuperscript{29,30} Although significant progress has been made, credited to their advanced capability and applicability, investigations on large area remain problematic due to their throughput restrictions. A classical throughput value for IBL is that writing 1 Tb of features takes roughly 1 month of continuous exposure time. In comparison, the physics involved with IL process are sufficiently straightforward. The principle is based on the interference between two beams, split from a coherent laser source, forms a standing wave that is recorded on a photoresist coated wafer at angles of $\theta$ and $-\theta$. The resulting interference pattern has a period of $\lambda/2\sin \theta$.\textsuperscript{1} For the light sources of LIL, lasers with wavelength that closely matches the photoresist, utilized for the semiconductor industries, have been the focus of the research. These wavelengths include 364 and 355 nm, which both match the I-line resists designed for 365 nm; doubled Ar at 244 nm, which matches KrF resists (248 nm); and 213 nm (fifth-harmonic of a Nd:YAG laser) and 193 nm (ArF laser), which match the industry-standard deep UV resists (developed for 193 nm). The minimal feature sizes possible are 182, 178, 122, and 96 nm, respectively. Further reductions in scale are possible using immersion techniques. Most of the effort on immersion IL to date has been at the ArF wavelength to generate minimal pattern sizes.

LIL is very innovative in nanolithography. This is mainly because of the higher efficiency compared to the EBL or IBL technology, and has a wide workspace and low cost. LIL has the following advantages compared with other nanolithography technologies: (1) low cost; (2) very high throughput; (3) no contamination on the surface; (4) capable of fabricating large-area patterns (up to hundreds of mm in diameter); (5) program controlled reconfigurable patterns (with different periods, feature sizes and pattern shapes).

2. LASER INTERFERENCE LITHOGRAPHY (LIL)

2.1. Process Parameters for LIL

Experimental characteristics could be defined by exploring the effects of several key parameters of LIL process, which could provide further understanding. These parameters include exposure dosage, beam power, and the half angle between beams. In addition, optimization of the generated patterns is of great importance, which could be achieved by the application of anti-reflective coating materials. To simplify the process, a single beam Lloyd’s mirror interferometer was considered for the experiments\textsuperscript{31} with 515-nm wavelength laser, frequency was doubled with a beta-barium borate (BBO) crystal.\textsuperscript{32} In order to define the required exposure dosage, case studies were performed with constant values of other parameters, such as LIL angle and beam power. It can be clearly seen that insufficient dosage resulted in holes (Fig. 1(c)) instead of columns, whereas excessive dosage generated inconsistent shape of patterns and could result in collapse of the structure (Figs. 1(d) and (e)), with the utilization of a positive photoresist.
Intensity distribution is another critical factor for fabricating uniform periodic patterns especially on large area, where it is a function of the wave amplitude of the partial beams, the wavelength of the light source, and the LIL angle. For a two-beam interference pattern, intensity $I(x)$ is given by:

$$I(x) = 2A \left\{ \cos \left( \frac{4\pi x}{\lambda} \sin(\theta) \right) + 1 \right\}$$

(1)

where $A$ is the wave amplitude of the partial beams, $\lambda$ is the wavelength, and $\theta$ is the LIL angle. Utilizing the spatial filter to expand the beam could minimize the non-uniform intensity distribution; however, for larger area fabrication, higher expansion of the beam or longer distance to the sample may be required.

Followed by the definition of the required energy dosage, the effects of the half angle between two beams at the intersection were explored by the rotation of the stage and thus varying the angle $\theta$. As mentioned previously, the relationship between the LIL angle ($\theta$) and the pitch of the pattern ($\Lambda$) can be expressed as:

$$\Lambda = \frac{\lambda}{2} \sin \theta$$

(2)

By observing the size of the pitch and the LIL angle, it is apparent that Figure 2 agrees well with the above equation, where the smallest size of the pitch is half the wavelength of the laser.

Moreover, the power of the beam could be increased to reduce the exposure time for quickening the LIL procedure. Although this seems to be apparent, since the total energy applied to the sample is identical, the difficulties in realizing large area uniformity could arise as the area of intensity distribution decreases.


Figure 2. (a)-(d) SEM images of transferred Si NM on glass substrate with light incident angles of 5, 10, 15, and 20 degrees, respectively. (e) Measured hole and pitch sizes as a function of the LIL angle. Reprinted with permission from [66], J.-H. Seo, et al., IEEE Photonics J. 5, 2200106 (2013). © 2013, IEEE.
2.2. Enhancement on Pattern Resolution

The gradual decrease in thickness of the nanoscale pillar, shown in Figure 1(e), is due to the vertical standing waves caused by the reflection from the bare silicon substrate.\(^\text{34}\)

To prevent generation of undesired patterns, anti-reflective coating is widely used where it controls the reflection of the laser light source at the surface of the substrate.\(^\text{35–38}\)

Figure 3 shows the comparison between samples with and without the application of bottom anti-reflective coating materials (BARC) for one- and two-dimensional patterns. The exposure dosage was kept constant, and identical experimental procedures were performed for each case. For the samples with the photoresist only, shown on the left side in Figure 3, inconsistent shapes could be observed. As the vertical standing waves were caused from the reflection from the Si substrate, the width of the pillar was continuously reduced as it reaches closer to the substrate and irregular wall surface could also be observed. By applying the BARC materials, improvements on the patterns could be clearly observed on right side of Figure 3.

With the introduction of the BARC materials, the patterns show more uniformity and stability, improved conditions for post processing, and a reduced number of inconsistent shapes and irregular wall surfaces preventing the possibilities of the collapse of the structures. Additionally, pattern footing and T-top phenomena from the development process were significantly reduced. Through application of the BARC materials, it is possible to generate smaller uniform patterns, enhancing the pattern resolution and size limitations of LIL.

Another important issue concerning LIL is clearly the pattern size limitation. Although short wavelength light source could be used and therefore minimize the generated feature sizes, further investigation on the reduction of pattern to pitch size ratio, less than half of the pitch length, could provide further understanding of the process.\(^\text{35}\)

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To reduce the pattern size by increasing the exposure energy, the LIL angle was set constant and the BARC materials were applied for uniform and stable pattern fabrication. Ar–Ion laser (257 nm wavelength) was utilized as the light source, and the pitch size was set to around 770 nm, where the energy doses varied from 9 to 18 mJ/cm\(^2\). Figure 4 shows the resulting top and cross-sectional views of the fabricated line patterns.

The measured pattern sizes are itemized in Table I. It could be clearly observed that increasing the exposure energy results in reduction of pattern size, reaching less than the quarter of the pitch size in this experiment. The inverse linear relationship between energy dose and the pattern size was also confirmed while maintaining the stability and uniformity of the patterns. These results

<table>
<thead>
<tr>
<th>Energy dose (mJ/cm(^2))</th>
<th>Pitch size (nm)</th>
<th>Pattern size (nm)</th>
<th>Ratio (pitch/pattern)</th>
</tr>
</thead>
<tbody>
<tr>
<td>9</td>
<td>~770</td>
<td>390</td>
<td>1.97:1</td>
</tr>
<tr>
<td>12</td>
<td>340</td>
<td>2.27:1</td>
<td></td>
</tr>
<tr>
<td>13.5</td>
<td>290</td>
<td>2.66:1</td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>190</td>
<td>4.05:1</td>
<td></td>
</tr>
</tbody>
</table>

highlight the possibility of pattern reduction that could be applied for various short wavelength light sources to generate minimal sized features and offer different geometrical designs for post procedures such as reactive ion etching and photoresist lift-off.

As mentioned above, LIL is a technique that involves a maskless process suitable for large-area patterning. Although this capability is one of the main advantages for most LIL applications, optional selective patterning is often required for many applications. To account for the non-selective patterning limitation of LIL, design and development of combined LIL and photolithography have been reported.39

Combined LIL and photolithography patterning technique consists of reactive ion etching in between the LIL and the photolithography process to fabricate a hybrid mask mold containing micro and nanopatterns. Introduction of a sacrificial layer is required for nanoscale patterns and of a photomask for microscale patterns.

The possibility of pattern resolution enhancement utilizing BARC, variation of exposure energy, and selective patterning could lead to various applications, especially for device production.

3. APPLICATIONS OF OPTICAL DEVICES BY LIL
3.1. Large-Area Photonic Applications
Two-dimensional (2D) photonic crystal slabs (PCSs) are one of the most promising artificial platforms for optical applications such as microcavities, lasers, photodetectors, solar cells, sensors, reconfigurable photonics, etc.30,41

Many conventional nanopatterning techniques involve a slow process and are not cost effective. LIL, on the other hand, enables generating periodic patterns faster on the larger area than conventional methods, and thus is suitable for creating 2D PCS structures. Given this, fabrication of PCSs with LIL suggests the easiest method to achieve large-size PCSs without losing performance. Most 2D PCSs were made by further deposition or etching to the substrate using photoresist patterns defined by LIL.42,43 Therefore, 2D PCSs by LIL should make either single crystalline substrate and form a homogeneous layer structure or amorphous/polycrystalline layer structure by deposition followed by lift-off. For the former case, it is difficult to take advantage from refractive index difference between the PCS layer and substrate and for the latter case, strong absorption loss, due to the defects in amorphous/polycrystalline material, often degrades overall device performance. As a result, this limitation hinders creating a more delicate and complex photonics system.

The recent advances of releasable and transfer-printable Si nanomembranes (NMs),44–53 still inheriting the single crystal quality of bulk Si, have enabled Si to be applicable to optical devices built on the foreign substrate by printing method.54–59 While there are many photonic crystals made with polymers, amorphous materials, and metals,60–65 Si NMs have very high index but the absorption loss is almost negligible; therefore, with Si NMs a unique and high performance 2D PCS can be realized. In this review, we introduce 2D Si NM PC mirrors built on the glass substrate by transfer printing method as an example.66

Figure 5 shows the typical fabrication process for 2D PCSs built on a glass substrate with Si NM. Fabrication begins with patterning nanostructures on the SOI wafer (SOITEC™). Top Si with the photoresist as etching mask was etched by deep reactive ion etching (DRIE), followed by the selective undercut etching of the buried oxide layer.

The released patterned top Si layer is, then, referred to as Si membrane reflector (Si-MR). This Si-MRs were picked up and printed onto the glass substrate by employing a polydimethylsiloxane (PDMS) stamp printing technique without using any adhesive layer between the Si-MR and the glass substrate.67 After transfer, the sample was annealed at 220 °C for 180 sec using RTA under nitrogen atmosphere to improve the bonding strength between the transferred Si-MR and the glass substrate. Images in Figure 6 show the finished Si-MR on SOI substrate.

Figures 7(a) and (b) show the SEM images taken on the Si-MR after printing on the glass substrate. The dimensions of nanopatterns are 240 nm hole radius with 500 nm pitch distance. Shown in Figures 7(c) and (d) are sample images of a 2 cm × 2 cm sized Si-MR, before and after printing onto the glass substrate. With the PDMS-assisted printing process, large-area and high-quality Si NMs have been successfully transferred without any visible fractures, and finally, Si-MR was fabricated on the foreign substrate (a glass substrate).

The reflectivity of the MR was measured at normal incidence with a beam size of around 100 μm. The spectra are shown in Figures 8(a) and (b), for the reflectors before printing (on SOI wafer) and after printing (on the glass), respectively. The reflections of these Si-MRs at different locations are also tested, and the results are plotted together using different colors.

As shown in Figure 8(a), the Si-MR peak reflectivity on SOI is as high as 95% at a near infrared wavelength range (∼1300 nm) with good uniformity at different locations. The bandwidth is about 100 nm from 1250 to 1350 nm. Theoretically, 100% reflection can be achieved, which can be realized with further fabrication optimizations. On the other hand, some have used LIL by forming a periodically structured array to decrease light reflection from the surface.68,75 Previously, patterning by e-beam lithography or spreading nanospheres followed by dry-etching are the most popular methods to generate a reasonably high anti-reflective coating layer.50–75

But LIL can be easily applied to fabricate large-area anti-reflective coating. Theoretically, reflection reduces as the layer thickness over the target wavelength increases.69


Figure 5. Schematical illustration of fabrication process for Si-MR on the glass substrate using transfer printing technique with assistant of elastomeric stamp.

Figure 6. Sample images after LIL pattern followed by RIE etching on SOI wafer.

Thus anti-reflection at certain wavelengths can be easily realized by the relationship shown in Figure 2. Hadobas et al. demonstrates anti-reflective coating layer on Si substrate and about 5% of reflection in visible range was achieved as shown in Figure 9.75

3.2. Plasmonic Color Filter

The “plasmonics” has garnered much interest by many scientists due to its interdisciplinary characteristics. The plasmonic structures accompany an optical resonance phenomenon, which is called surface plasmon resonance (SPR), the free electrons oscillate at the surface of metal. Researchers have been amassing the ways to handle the metal in nanoscale structures with various nanofabrication methods, such as e-beam lithography, ion milling, self-assembly, interference lithography, and so on. That has led to advances in research approaches with the SPR by suggesting various applications.76–81

The color filters widely used for the industrial devices such as organic light emitting diodes (OLEDs), Liquid Crystal Displays (LCDs), and CMOS image sensors, are composed of organic dyes (or pigments). The color-reproduction performances of these dye-based filters are highly dependent on the material intrinsic characteristics; color resist in the red filter, for example, absorbs all wavelength regions except the light in red range. The filtering performance originated from color sensitivity of dyes is degraded by heat and ultraviolet radiation due to low chemical stability of the organic materials.82–83

On the other hand, the PCFs are combining on an optically thin metal layer, and its transmittance can be tuned by the geometrical and material conditions: the periodicity, size and shape of holes, the thickness of metal, and the optical constants of materials. This simple and thin structure is advantageous to be assembled into other devices regardless of degradation by heat and light. Many reported results in earlier stages have demonstrated PCFs as indicating easy tunability of transmittance.84–87 More recently, experimental analysis about spatial cross talk and the effect of defects was done in detail, and showed more possibility to industrial applications.79
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Figure 10. Schematic diagram of the proposed fabrication process. In a Lloyd’s mirror interferometer system, two laser beams form a 1D interference pattern on the specimen. To create array patterns with a square symmetry, two exposures by a rotation of 90° in between are applied. Reprinted with permission from [102], Y. S. Do, et al., Adv. Opt. Mater. 1, 133 (2013). © 2013, Wiley-VCH.

Table II. The dimensions of the optimized structure for each color. The average values of the diameter of the holes in the fabricated samples were rounded off to the tens place.

<table>
<thead>
<tr>
<th>Color</th>
<th>Period (p) [nm]</th>
<th>Thickness of Al [nm]</th>
<th>Diameter of holes [nm]</th>
<th>Diameter of holes (experimental) [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Red</td>
<td>390</td>
<td>150</td>
<td>200–230</td>
<td>~210</td>
</tr>
<tr>
<td>Green</td>
<td>320</td>
<td>150</td>
<td>180–200</td>
<td>~180</td>
</tr>
<tr>
<td>Blue</td>
<td>230</td>
<td>100</td>
<td>120–160</td>
<td>~120</td>
</tr>
</tbody>
</table>


However, the fabrication method used until now, such as nanoimprinting84,88–90 electron beam lithography,76–81 or focused ion beam,91–95 restricts mass-production of PCFs, encountering problems of low speed, small patterning area, and high cost equipment. Here we suggest a fabrication flow including laser interference lithography (LIL) step. As opposed to above-mentioned technologies, LIL yields perfect ordering patterns, which must be spatially coherent over large area, with simple maskless equipment. Although LIL has the limitation that it can only fabricate simple periodic patterns, it is an attractive additional solution of conventional methods for applications in which periodic patterns are desirable, including X-ray transmission gratings, photonic crystals, and sub-micrometric sieves. In this regard, fabrication of plasmonic color filters with LIL is the easiest method to achieve large size PCFs without losing performance aspect. In this work, we demonstrate PCFs with primary colors (red, green, and blue) on a 2.5 cm × 2.5 cm area. The transmittance of the filters was optimized by analyzing SP modes at the interfaces between Aluminum (Al) and Lithium Florid (LiF).

Figure 10 shows the suggested fabrication procedure. A LiF layer (50 nm) and an Al layer (150 nm for the red

Figure 11. Transmittance of the PCF of which the cylindrical surface as well as the top surface are covered with LiF of (a) 100 nm and (e) 200 nm, respectively. (b)–(d) Illustrate the electric field intensity, \( \log(\bar{E}^2) \), at the wavelength of \( \lambda_1 \), \( \lambda_2 \), and \( \lambda_3 \) in (a). (f)–(h) Illustrate the electric field intensity, \( \log(\bar{E}^2) \), at the wavelength of \( \lambda_1 \), \( \lambda_2 \), and \( \lambda_3 \) in (e). Reprinted with permission from [102], Y. S. Do, et al., Adv. Opt. Mater. 1, 133 (2013). © 2013, Wiley-VCH.
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Figure 12. Transmittance of the PCFs according to the thickness of the LiF layer encountering the bottom of the Al film.

and green; and 100 nm for the blue) were thermally evaporated in sequence on the glass substrate. In the following, laser interference lithography was performed with a Lloyd's mirror interferometer system. To make the interference patterns, two beams were used: one travels directly to the specimen and the other was reflected onto the specimen by the mirror. These two beams formed a 1D interference pattern on the specimen. The optimized dimensions for each structure are specified in Table II. The last column indicates the average hole size, which was patterned on the photoresist. The size of the hole can be tuned by adjusting the exposure dosage, the beam power, and the time for developing. In order to optimize the transmission characteristics, structures consisting of a unit cell were simulated by a three-dimensional (3D) finite-difference, time-domain method (FDTD, Lumerical solutions, Canada). The optical constants of the glass substrate and the Al were based on the Palik data; the optical constant of LiF was determined from the experimental data. A plane wave source polarized in the x direction was launched at normal incidence.

As shown in Figure 11(b), the $SP_{b,1}$ and $SP_{u,1}$ modes occurred simultaneously at $\lambda_3$, similar to the $SP_{b,0}$ and $SP_{u,0}$ modes at $\lambda_1$ (Fig. 11(d)), resulting in a similar amount of strong field distribution at the top and bottom of the hole. The $SP_u$ mode at $\lambda_2$ appeared to be much weaker than it was at $\lambda_3$ and $\lambda_1$. We supposed that the top of the structure became planar when the upper LiF was deposited in a layer more than 150 nm thick. Figures 11(e)–(h) illustrates the response of the structure with the upper LiF layer of 200 nm, indicating that the matched $SP_b$ and $SP_u$ modes were maintained after planarization.

The thickness of the LiF at the bottom of the metal-dielectric interface had a relatively small effect on the $SP_b$ modes. Because the thickness of the LiF was scaled to the penetration depth of the surface plasmon ($\delta_2$), the thickness affected the effective permittivity ($\varepsilon'_d$), which ranged from the permittivity of: (1) the air to LiF in the case of the upper LiF and (2) LiF to SiO$_2$ for the bottom. The difference in permittivity between LiF and SiO$_2$ was less than that between LiF and the air: 0.23 for the former one; 0.93 for the latter, at a 400 nm wavelength. Therefore, the LiF between the substrate and Al had less influence on the transmittance, as shown in Figure 12.

Figure 13(a) provides the photograph of the PCFs. The color gamut of each filter was mapped on the CIE 1931 \textit{x}y chromaticity diagram (Fig. 13(b)). As with the transmittance results, the red and blue filters showed worse color purity compared to that of the green filter. Since $Z$, which is quasi-equal to the blue stimulation, is the biggest among the tristimulus values, the blue-noise light can have the worst effect on the color purity. In the hexagonal arrays, the transmittance peaks originating from the 1st and 2nd...
resonance modes are split into further positions in wavelength region than are the square arrays. 99–101 This would be advantageous for producing pure red color. Fortunately, three laser beams make a 2D-interference pattern with a hexagonal array, so there is a chance, experimentally, to improve the color purity.

4. CONCLUSION

Among various nanoscale patterning techniques, LIL could be advantageous for its maskless, quick, and simple process characteristics. By exploring the experimental aspects of LIL, the advantages and limitations are highlighted for generating nanoscale patterns. Definitions and the effects of the critical process parameters, such as exposure dosage, intensity distribution, LIL angle, and beam power, have been discussed followed by pattern resolution improvement using anti-reflective coating materials. Minimizing the pattern size through variation of exposure energy and introduction of combined LIL and photolithography technique to account for non-selective patterning limitation of LIL has been discussed.

Optical/photonics applications by LIL indicate that simple, fast, and large area processable devices can be realized. We have reviewed various periodic structures such as reflectors and anti-reflectors where reflection is over 95% at a near infrared wavelength range for reflectors and is less than 5% for anti-reflective coating layers, respectively. This implies LIL can produce inexpensive key components in optical systems. We have also reviewed large area PCFs that are effectively colored as R, G, B. The results show great potential of PCFs to be applicable to display applications. The LIL process provides nano-scaled array patterns throughout the entire 2.5 cm × 2.5 cm area and obtained 20.0%, 19.9%, and 22.9% for the maximum transmittance of the red, green, and blue, respectively.

We expect that these optical/photonics applications will provide the opportunity to lead more advanced applications to mass production.

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References and Notes
