Adaptive optics enhanced direct laser writing of high refractive index gyroid photonic crystals in chalcogenide glass

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Abstract: Chiral gyroid photonic crystals are fabricated in the high refractive index chalcogenide glass arsenic trisulfide with an adaptive optics enhanced direct laser writing system. The severe spherical aberration imparted when focusing into the arsenic trisulfide is mitigated with a defocus decoupled aberration compensation technique that reduces the level of aberration that must be compensated by over an order of magnitude. The fabricated gyroids are shown to have excellent uniformity after our adaptive optics method is employed, and the transmission spectra of the gyroids are shown to have good agreement with numerical simulations that are based on a uniform and diffraction limited fabrication resolution.

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The chalcogenide glass arsenic trisulfide (As$_2$S$_3$) is a useful material for three-dimensional (3D) nanofabrication via direct laser writing (DLW) due to the excellent photo-polymerizability of its evaporatively deposited thick films [1–4]. The high refractive index [5], good transparency in the near- to mid-infrared wavelength range [6], and the large third order Kerr nonlinearity [7] of As$_2$S$_3$ also make it a worthy candidate for the fabrication of photonic devices [8] including 3D variants such as mid-infrared waveguides [9] and photonic crystals (PhCs) [1–4].

However, DLW in As$_2$S$_3$ with a high numerical aperture (NA = 1.4) objective lens is often hindered by the so called refractive-index mismatch aberration that is imparted onto a focused laser beam when it passes through the large mismatch of refractive indices between the As$_2$S$_3$ ($n = 2.35$) and the immersion oil ($n = 1.52$) of the objective lens [10, 11]. The extreme magnitude of the aberration in combination with its linear dependence on the focal depth [10–12] can impart significant spherical aberration and defocus onto the focused laser beam, which, by causing a drop in the peak intensity of the focus below the fabrication threshold, can ultimately lead to a complete cessation of fabrication [12–14]. Moreover, as the depth dependence of the aberration imparts a depth dependence on the peak intensity of the focus, a strong asymmetry in the fabrication feature size is imparted onto the fabricated nanostructure such that mechanical stability and uniformity are lost. For photonic nanostructures such as photonic crystals (PhCs) [15] that demand a high degree of uniformity, such a depth dependent asymmetry is a severe hindrance to performance [14].

In media that have lower refractive indices with respect to As$_2$S$_3$, adaptive optics shows promise in compensating for the refractive-index mismatch aberration [16–18]. However, many adaptive optics techniques that rely on spatial light modulators (SLMs) have not been applied to the compensation of the large magnitude refractive-index mismatch aberration in the DLW of...
complicated 3D micro-/nano-structures [19]. In addition, some compensation processes place unnecessary strain on the adaptive optics element by compensating for the full refractive-index mismatch aberration function [12]. Other techniques that reduce the strain by compensating for only the spherical aberration component can be hindered by the need for pre-computation or measurement of the actual focal location in order to place the focus at the desired position [14].

Here we present the fabrication of high refractive index chiral gyroid PhCs [20–23] in As$_2$S$_3$ via adaptive optics enhanced DLW. These complex and fully interconnected 3D nanostructures demand a high degree of uniformity in order to produce strong and consistent circular dichroism. The gyroid also forms the basis of a number of more complex chiral geometries [21] that require the large refractive-index contrasts that As$_2$S$_3$ offers. Thus, due to the asymmetry that would otherwise be imparted by the refractive-index mismatch aberration, As$_2$S$_3$ gyroid PhCs fabricated by means of DLW require stringent adaptive aberration compensation. To this end, we develop a powerful adaptive aberration compensation technique to correct for the refractive-index mismatch aberration during the DLW of As$_2$S$_3$. Our technique, which incorporates an efficient and optimal defocus decoupling scheme is shown to reduce the magnitude of the refractive-index mismatch aberration compensation by over an order of magnitude, enabling optimal and dynamic aberration compensation during vertical translations. We show through circular polarization sensitive transmission spectroscopy that the circular dichroism of the fabricated gyroid PhCs is in good agreement with numerical simulations that are based on a uniform and diffraction limited fabrication resolution.

2. Adaptive aberration compensation for the DLW of As$_2$S$_3$ gyroid PhCs

We will here use the term ‘gyroid PhC’ to designate a PhC based on the single gyroid geometry [20]. The single gyroid, also termed srs-net, is a three-valent network with cubic symmetry and spatial chirality that manifests in identical four-fold screw rotations in all coordinate directions and three-fold screw rotations (of opposite handedness) along the body-diagonal directions [20, 21]. The 3D connectivity of the srs-net, which can be seen in the 3D and 2D diagram of a left-handed (LHD) simple cubic unit cell in Fig. 1, necessitates vertical translations throughout the fabrication medium when the network is replicated via DLW. As the focal depth is altered during such translations, the magnitude of the refractive-index mismatch aberration will also be altered [10–12] and thus, an adaptive optics compensation scheme is required to ensure diffraction limited DLW [12].

![Fig. 1. The gyroid PhC simple cubic unit cell.](image_url)

Here we present a powerful defocus decoupled adaptive aberration compensation scheme that optimizes and simplifies the aberration compensation process for application to DLW. The full derivation of our compensation scheme can be found in Appendix A, but the process can be visualised here as the simultaneous combination of the following three components. One,
the geometrical focal depth, \(d_0\), of the objective lens is positioned axially such that it relates to the desired fabrication location, \(d_1\), via the relation \(d_0 = sd_1\). The parameter \(s\) is hereafter referred to as the defocus factor and can be visualized in the diagram of the focusing geometry in Fig. 2(a) for the case where \(s < 1\). Two, the spherical aberration and defocus that is present when focusing to a depth of \(d_0\) is compensated by applying the conjugate of the refractive-index mismatch aberration function [10, 11] to the SLM. The position of the actual focus after this step coincides with the geometrical focal depth at \(d_0\). Three, the position of the focus of the objective is shifted axially from \(d_0\) to \(d_1\) through the subtraction of a spherical defocus function from the refractive-index mismatch aberration function.

![Diagram](image1)

Fig. 2. (a) Defocus decoupled focusing geometry. (b) Plot of the peak to peak value of \(\psi'_R\) as a function of \(s\) for \(d_1 = 20 \mu m\). The five images within the plot are phase wrapped images of \(\psi'_R\) when \(s = 0, 0.25, 0.498, 0.75\) and \(1\).

Under this defocus decoupling scheme, the final phase compensation patterns for a diffraction limited focus at \(d_1\) are found by combining the phase from components two and three. The combination yields the equation (see Appendix A for derivation)

\[
\psi'_R(\rho) = k_0d_1 \left[ s \sqrt{n_1^2 - NA^2\rho^2} - \sqrt{n_2^2 - NA^2\rho^2} \right],
\]

where \(k_0\) is the wavevector of the wavefront in vacuum and \(n_1\) and \(n_2\) are the refractive indices of the first and second medium, respectively, and \(\rho\) is the normalized pupil radius.

![Diagram](image2)

Fig. 3. Five refractive-index mismatch aberration compensation patterns at fabrication depths of 0, 3, 6, 9 and 12 \(\mu m\) when (a) \(s = 1\) (i.e. no defocus decoupling) and (b) \(s = 0.498\) (i.e. optimised defocus decoupling).
The advantage of defining the compensation scheme in this manner is that the level of defocus removal can be optimized to minimize either the magnitude or gradient of the phase displayed on the SLM. Figure 2(b) shows a plot of the optimization of the peak to peak magnitude of $\psi'_R$ as a function of $s$ for the case of $NA = 1.4$, $n_1 = 1.52$, $n_2 = 2.35$, $d_1 = 20 \mu m$ and $k_0 = 2\pi/\lambda$ where $\lambda = 800$ nm. Also shown in Fig. 2(b) are five 0 to $2\pi$ phase wrapped images of $\psi'_R$ for various defocus factors. At $s = 0.498$, a clear minimum in the peak to peak magnitude is observed, corresponding to over an order of magnitude reduction in phase that must be displayed on the SLM and a subsequent reduction in the number of 0 to $2\pi$ phase wraps from 11 to 0. It should be noted that whilst this numerical optimization is trivial, it is also possible to derive an analytical solution for $s$ as we show in Appendix B.

The effect of the reduced phase magnitude on the compensation patterns needed for even modestly sized $\text{As}_2\text{S}_3$ gyroids can be observed in Fig. 3 for the case of a 12 $\mu m$ tall gyroid. The phase patterns in Fig. 3(a), which are created when $s = 1$, (i.e. $d_0 = d_1$), have a larger number of phase wraps in comparison to the phase patterns in Fig. 3(b) where $s = 0.498$ (i.e. $d_0 \approx d_1/2$).

3. Fabrication of $\text{As}_2\text{S}_3$ gyroid PhCs

Adaptive optics enhanced nanofabrication of $\text{As}_2\text{S}_3$ gyroid PhCs was performed via the DLW of 16 $\mu m$ thick films of evaporatively deposited $\text{As}_2\text{S}_3$ [2] using the experimental setup described in Appendix C. The lattice constant of the fabricated gyroids was 3 $\mu m$ and the total gyroid dimensions were 4x4x4 unit cells. Gyroid PhCs were fabricated both without any adaptive aberration compensation, and with the SLM adaptively compensating for both system aberration and the defocus decoupled refractive-index mismatch aberration function from Eq. (1). The laser power was varied within the range of 2 to 3 whilst the fabrication speed was constant at 10 $\mu m/s$ for fabrication close to the threshold of $\text{As}_2\text{S}_3$. After fabrication, the $\text{As}_2\text{S}_3$ film was chemically etched [1–4] to reveal the free-standing gyroids.
be seen in Figs. 4(b) and 4(d), respectively. The axial and lateral feature sizes for each of these cases are plotted in Figs. 4(e) and 4(f), respectively. As a result of the drop in peak intensity as the focal depth increases [12, 13], a loss in uniformity occurs for the uncompensated gyroids with the feature sizes furthest from the glass substrate (depth as labelled) being smaller than the layers close to the substrate. In contrast, the use of defocus decoupled adaptive aberration compensation shows that symmetry can be maintained throughout the structure as indicated by the constant axial and lateral feature sizes at the depth is increased.

It should be noted that the increase in feature size once aberration compensation is employed is not due to a broadening of the point spread function that occurs under the presence of spherical aberration, but arises from the fact that the peak intensity of a diffraction limited focus is larger than the aberrated cases such that a greater proportion of the point spread function lies above the fabrication threshold. It should also be noted that whilst the axial and lateral dimensions of the point spread function do broaden under the presence of spherical aberration, the asymmetry introduced by these increases is dwarfed by the large drop in feature size that occurs from the thresholding process.

4. Transmission spectra and circular dichroism

The true benefit of adaptive optics enhanced DLW is evident in the transmission spectra of the gyroids. Figures 5(a) and 5(b) show the measured transmission spectra for right-handed circular polarization (RCP) and left-handed circular polarization (LCP), respectively, for a LHD gyroid with a 3 µm lattice constant and a size of 33x33x4 unit cells that was fabricated with the full defocus decoupled adaptive aberration compensation scheme. The fabrication power for the gyroid was set at 2.6 µW whilst the fabrication speed was set at 10 µm/s. Each experimental measurement was made with a Fourier transform infra-red (FTIR) microscope and spectrometer fitted with a broadband linear polarizer and a quarter-waveplate as well as a pinhole that limited the measurement to a ±5° (NA < 0.1) cone of light around the gyroids [001] direction. Figures 5(c) and 5(d) show the corresponding simulated transmission spectra for a gyroid with identical properties to the fabricated structure, such as the measured lateral and axial feature sizes of 195 nm and 975 nm, respectively, which were measured via the SEM images shown in Figs. 5(e) and 5(f). Full details of the simulation can be found in Appendix D.

Fig. 5. (a) Measured RCP, (b) Measured LCP, (c) Simulated RCP and (d) Simulated LCP transmission spectra along the [001] direction of a gyroid with a size of 33x33x4 unit cells. (e) and (f) are SEM images of the measured structure which were used to determine the lateral and axial feature sizes for the simulations.
For the case of the measured RCP spectra, a primary band of transmission suppression can be seen at a wavelength of 3.65 µm, precisely as predicted by the numerical simulations. This strong agreement in the spectral location, which also exists for the other features of the RCP spectra as indicated by the red arrows between the plots, is evidence that the position of the focus is being precisely controlled by our defocus decoupled adaptive aberration compensation scheme. Should the defocus decoupling mechanism have been under or over compensating the position of the focus, then the lattice constant of the gyroid in the [001] direction would have been compressed or stretched, resulting in a blue- or red-shift, respectively, in both the measured LCP and RCP spectra.

The measured spectra also hold evidence of our compensation schemes ability to maintain uniformity. The gradient of the low-frequency RCP band-edge at 3.75 µm (0.846 %/nm) shows strong agreement with the numerical simulations (0.853 %/nm) that assume a completely uniform gyroid structure. Should a loss of periodicity have been introduced by the spherical aberration induced asymmetry, then the gradient of the band-edge would be expected to decrease given the diminishing constructive interference from the PhC.

Similar agreement can be found between the measured LCP spectra with respect to the simulated case. Whilst there is an evident blue-shift in the position of the measured spectra relative to the simulated spectra which has been in previously reported comparisons between the measured and simulated transmission spectra of laser written polymeric gyroids [22, 23], the width ($\Delta \lambda = 320$ nm at $T = 25\%$) and maximum strength ($T = 6\%$) of the primary band gaps agree well with the width ($\Delta \lambda = 358$ nm at $T = 25\%$) and maximum strength ($T = 1\%$) of the simulated structure.

Finally, additional evidence of the improvement that adaptive optics offers is seen in the comparison between aberrated and unaberrated transmission spectra in Fig. 6. The measured transmission spectra of gyroids fabricated without our adaptive compensation scheme can be seen in Fig. 6(a) whilst the transmission spectra of gyroids fabricated with our defocus decoupled adaptive compensation can be seen in Fig. 6(b). For each case, the fabrication power was varied around the threshold level of 2-4 µW whilst the fabrication speed was kept constant at 10 µm/s.

For the aberrated case, the transmission spectra are seen to behave turbulently as the fabrication power is increased around the fabrication threshold. This is because of rapid changes in the...
structural integrity and feature size when the refractive-index mismatch aberration influences the thresholding process. Across the entire 3.2 to 3.9 µm wavelength range of the spectra, inconsistent bands of circular dichroism are observed at near arbitrary locations. The maximum positive dichroism (TLCP > TRCP) of the uncompensated structures is seen to vary considerably with an average strength of +47±12 % whilst the maximum negative dichroism (TLCP < TRCP) is extremely variable and in some cases nonexistent with an average strength of -11±15 %. In contrast, the completely compensated gyroids show bands of circular dichroism with consistent spectral position and strength. A band of positive circular dichroism is observed to red-shift through the 3.2 to 3.5 µm wavelength range with an average strength of +66±2 % whilst a band of negative circular dichroism is observed to red-shift through the 3.5 to 3.7 µm wavelength range with an average strength of -57±4 %.

5. Conclusion

We have reported a defocus decoupled adaptive optics approach to substantially improve the spatial fidelity and uniformity of complex 3D nanostructures fabricated by means of DLW in the high refractive-index chalcogenide glass As2S3. We have shown that in 3D structures such as chiral gyroid PhCs that are sensitive to large-scale structural uniformity that our adaptive compensation scheme can produce uniform PhCs that have a pronounced circular dichroism feature in good agreement with numerical simulations that are based on a uniform and diffraction limited fabrication resolution. Such a compensation scheme will play a critical role in realising more complex chiral geometries [21] that demand large refractive-index contrasts.

Appendix

A. Defocus decoupled adaptive aberration compensation scheme

Here we derive our aberration compensation scheme that optimizes and simplifies the aberration compensation process for application to DLW. First, we recall that the geometrical focal depth, \( d_0 \), of our objective lens is positioned axially such that it relates to the desired fabrication location, \( d_1 \), via the equation

\[
d_0 = sd_1
\]

and that the spherical aberration and defocus that is present when focusing to a depth of \( d_0 \) is compensated by applying the conjugate of the refractive-index mismatch aberration function [10, 11] to the SLM. In normalized pupil coordinates, the conjugated function, \( \psi_R \), takes the form

\[
\psi_R(\rho) = -k_0 d_0 \left[ \sqrt{n_2^2 - NA^2 \rho^2} - \sqrt{n_1^2 - NA^2 \rho^2} \right], \quad (3)
\]

where \( k_0 \) is the wavevector of the wavefront in vacuum and \( d_0 \) is the geometrical focal depth. \( n_1 \) and \( n_2 \) are the refractive indices of the immersion and fabrication medium, respectively, whilst \( \rho \) is the normalized pupil radius related to the convergence angle (\( \theta_1 \)) in the immersion medium through the relation \( \rho = n_1 \sin \theta_1 / NA \).

In order to shift the position of the focus of the objective from \( d_0 \) to \( d_1 \) and to simultaneously reduce the magnitude of the phase compensation, we subtract a spherical defocus function \( \psi_D \) from \( \psi_R \). The spherical defocus function can be written as

\[
\psi_D(\rho) = k_0 d_1 (1 - s) \sqrt{n_2^2 - NA^2 \rho^2}, \quad (4)
\]

where \( d_1 (1 - s) \) is the distance that the focus is shifted as shown in Fig. 2(a). This defocus function effectively alters the focal length of the objective lens by altering the radius of curvature of the objective’s wavefront.
After the subtraction of $\psi_D$ and with the help of Eq. (2), we arrive at the final form of our refractive-index mismatch aberration compensation, $\psi_R'$

$$\psi_R'(\rho) = \psi_R(\rho) - \psi_D(\rho) = k_0 d_1 \left[ s \sqrt{n_1^2 - NA^2 \rho^2} - \sqrt{n_2^2 - NA^2 \rho^2} \right],$$  \hspace{1cm} (5)$$

which importantly remains linearly proportional to $d_1$ such that the optimisation of $s$ will be independent of focal depth and hence the structure height.

### B. Analytical solution for the case of optimized peak to peak phase magnitude

For the case of optimized peak to peak phase magnitude, the optimal value of $s$ corresponds to the condition when the phase magnitude at the centre and edge of the pupil are equal, i.e. when $\psi_R'(0) = \psi_R'(1)$. It is therefore possible to derive an analytical expression for $s$ that can be used to further simplify the compensation process. After some routine calculation, $s$ is revealed as

$$s = \frac{n_2 - \sqrt{n_2^2 - NA^2}}{n_1 - \sqrt{n_1^2 - NA^2}},$$ \hspace{1cm} (6)$$

which importantly varies only on the parameters of $NA$, $n_1$ and $n_2$ such that $s$ itself is constant for the purpose of DLW. For the parameters used in this work ($NA = 1.4$, $n_1 = 1.52$ and $n_2 = 2.35$), we find $s = 0.498$ in agreement with our numerical optimization.

### C. Experimental setup for adaptive optics enhanced DLW

A diagram of our experimental setup can be seen in Fig. 7. A Shack-Hartmann wavefront sensor (HASO3-128-GE, Imagine Optic) is used to measure system aberrations whilst a liquid crystal based SLM (HSPDM5120785-PCle, Boulder Nonlinear Systems) is used for simultaneous compensation of both the system and refractive-index mismatch aberrations. A photo multiplier tube (PMT) is used for detection of the As$_2$S$_3$ surface as well as for feedback on the refractive-index mismatch aberration compensation [12]. The laser is an amplified Ti:Sapphire laser system (Libra HE, Coherent Scientific) and is expanded to uniformly illuminate the surface of the SLM.

![Diagram of experimental setup for adaptive optics enhanced DLW](image)

Fig. 7. Experimental setup for adaptive optics enhanced DLW.

To ensure that only phase modulated light was used during fabrication, a 0 to $60\pi$ phase ramp was added to the aberration compensation phase such that the reflected diffraction orders of the SLM separated. The first diffraction order was imaged along the optical axis of the system to the
back aperture of the objective lens (Olympus UPLANSAPO 100XO) by a pair of achromatic lenses in a 4f configuration whilst a pinhole with a diameter of 1 mm was placed at the first Fourier plane so that the zero and higher diffraction orders were blocked.

D. Numerical simulations

The numerical simulations were performed using the commercial finite element software CST Microwave Studio. Each simulated gyroid was modelled as a lossless dielectric that had a refractive index equal to that of bulk As$_2$S$_3$ [5] whilst the feature sizes of the simulated gyroids were restricted to the measured axial and lateral feature sizes that were determined by SEM imaging of the fabricated structures. The height of the simulated gyroids matched the number of unit cells of the fabricated structures, whilst in the in-plane directions periodic boundary conditions were used for plane wave illumination. For accurate comparison to the experimental transmission spectra that were taken under low NA (< 0.1) rather than plane wave conditions, a total of 57 simulations were performed over an equivalent range of incident angles to the experimental case (±5°). The resulting spectra were averaged and smoothed to yield a close approximation to the continuous range of incident angles that are excited in the experiment.

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