Phase and Absorption Metrology for Thick Photopolymer Devices

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ABSTRACT

Studies of development kinetics in volume photopolymers typically use transmission holography to quantify the index distribution. This method has advantages including simplicity, quantitative index data and natural mapping onto theories using harmonic expansion of the material response. A particular disadvantage is that the low spatial-frequency response corresponding to the intensity of the writing beams can never be Bragg matched and thus remains invisible.

In configurations where the exposure is not primarily sinusoidal, the holographic method is not applicable. Important examples include bit-oriented data storage, direct-write lithography, and the object beam of page-based holography. In these cases the exposure intensity is essentially arbitrary and there is a need for metrology tools that can quantitatively measure the real and imaginary parts of the weak 3D index perturbation. Images produced by bright-field and phase-contrast microscopes are generally not quantitative and are corrupted by objects out of the focal plane.

We have developed two methods, a form of optical diffraction tomography and a scanning transmission microscope, that are specifically designed to measure the 3D index response of holographic materials. Both are optimized to measure the extremely weak absorption and phase structures typical of photopolymers and have passbands that match the expected spatial frequencies.

Keywords: Photopolymers, Volume Holography, Phase-Contrast Microscopy, Optical Diffraction Tomography

1. INTRODUCTION

Models of volume holographic photopolymer development \cite{1,2,3,4} have traditionally been based on sinusoidal excitation because of the relevance to holography but also because the model predictions can be easily validated by reading out a hologram and its harmonics. However, this restricts the metrology to the rather narrow range of spatial frequencies that can be Bragg matched. For example, the so-called “ambiguity” term which is recorded by the intensity of the object beam is not revealed. The profile of this low spatial frequency object is important both to validate models and due to its technical importance as an aberration or noise source in holographic data storage (HDS).

Similarly, in micro-holographic bit-wise volume data storage \cite{5}, diffraction efficiency decreases as the focus of the read head is offset in any direction from the bit. While this provides some measure of the size of the feature via the spatial overlap of the optical intensity and the index profile, Bragg matching also impacts the spatial dependence of efficiency. For example, a hologram with uniform amplitude and Gaussian phase profile extending throughout the depth of the material would result in a diffraction profile versus the depth of the read focus that is strongly localized in depth, totally unlike the actual uniform amplitude profile of the hologram.

Applications outside holography for these materials such as guided-wave optics written either by mask lithography \cite{6}, through launch of writing light from the core of an embedded fiber \cite{7}, or by direct-write lithography \cite{8} all create \textasciitilde10 \textmu m or larger features that can not be measured via holographic diffraction. Since the waveguides are

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typically deeply buried, common waveguide measurement methods such as prism coupling can not be applied. While some information is revealed by the size and shape of the guided modes, these are not unique.

The common feature of these examples is that the “signal” is only a portion of the story. The read channel in a data storage system or the guided mode of a waveguide does not reveal critical features of the index profile. To develop and validate holographic materials response models as well as to fully optimize and engineer applications of these materials, new forms of metrology are needed.

Existing bright-field and phase-contrast microscopes unfortunately do not meet this need. For traditional transmission microscopy, out-of-focus objects are superimposed on the image at the focus. If the point spread function of these microscopes is integrated transversely, conservation of energy forces the result to be independent of depth [9]. This proves that signals generated at all planes are equally present in the image, superimposed with different transfer functions. This makes quantitative interpretation of 3D objects very difficult. It is possible to deconvolve the point spread function (PSF) to produce true 3D images [10] but the penalty is generally loss of signal to noise ratio (SNR) which is not appropriate for the very weak objects under consideration here.

The out-of-focus planes can be physically rejected by microtoming the sample into thin (1 to 10 µm) slices. Unfortunately, the trade-off of resolution and depth of field reemerges here. For micron-scale resolution in the visible, the sample thickness has to be only a few microns. For a weak phase object of \( \delta n = 10^{-4} \), this provides much less than 1/1000 waves of delay which is insufficient to be detected.

Finally, unless one is using computational deconvolution, the transfer function of most phase-contrast microscopes is not constant across the passband or worse contains significant voids. A high-pass filter characteristic may be ideal for sharpening biological images, but it attenuates critical information in a metrology application. In many phase-contrast microscopes, amplitude and phase in the object are mapped into a single channel, further complicating the ability to quantify the result.

Thus the ideal system would provide true 3D information while rejecting out-of-focus signals, have a small point spread function and flat passband for distortion-free high resolution imaging, return independent quantitative amplitude and phase information and include some form of gain to improve the SNR of weak objects. We introduce two methods here that meet many of these criteria and compare them to a standard phase-contrast microscope.

2. BACKGROUND

As a concrete example, consider the recording of holograms in plane-wave multiplexed HDS. For sake of simplicity, consider the object beam to be a high numerical aperture (NA) cone; in reality, modulation within this cone represents the information to be stored. Assuming a material with a perfectly linear response, this object records an index feature proportional to its intensity, which in Fourier space is contained within the double leaf shape, as shown in Figure 1. If no phase mask is placed on the spatial light modulator of the HDS system, the DC content of the object appears in the Fourier plane as a spike at the origin. Most measurement techniques can not read this spike, resulting in a measurement of \( \delta n \) but not absolute index. The holograms recorded as the angle of the reference beam is discretely stepped across its range are shown as lines in the figure. Individual pixels of the SLM are positioned along this line. For sake of simplicity, we will restrict this discussion to 2D, although in reality the Fourier-space diagrams extend into the page.

Although we are using the example of HDS, note that any “object” focused through the polymer records a pattern similar to the red cross-hatched object in Figure 1. Direct-write lithography with a focused beam [8] or bit-wise data storage via index “bumps” [11] would thus occupy an identical region, while adding a counter-propagating beam to write micro-holograms [5, 12] simply shifts the pattern in Fourier space via the carrier frequency to \( k_z = \pm 2k \). The discussion can thus be easily generalized to other applications.

We can now examine potential microscopes by comparing their transfer functions to the spatial frequency content of the object we wish to measure. The various options are summarized in Figure 1,
Confocal reflection microscopy (CRM) is the gold standard of 3D microscopes. As shown in Figure 2, a focused beam is translated through the sample and the magnitude of the reflection is detected after filtering by a pinhole at the image of the focus. This pinhole rejects out-of-focus signals enabling true 3D mapping. However, for the applications considered here, there are several problems. First is that the transfer function of the CRM does not overlap with the spatial frequencies of objects written into a linear material via transmission [13], even using an oil immersion objective to obtain the maximum NA equal to the polymer index. Another limitation is that the CRM mixes returns from amplitude and phase onto a single detector and thus does not provide independent information.

Scanning or static phase-contrast and bright-field microscopes have a variety of transfer functions depending on implementation, but all transmission systems share the region of support shown as a dashed line in Figure 1. Although they have the limitations mentioned earlier, if the NA of the microscope exceeds that used to write the object, the complete object can in principle be recovered. The first system considered will thus be a scanning transmission microscope that meets many of our stated objectives.

Finally, we will consider optical diffraction tomography. As shown on the left of Figure 1, when used to measure a planar sample in air, its transfer function has the same region of support as a transmission microscope of equal NA. However, it has the distinct advantage of independent real and imaginary channels with completely uniform passbands. Unlike most microscopy in which resolution must be traded against depth of field and working distance, ODT is unlimited in both, a critical feature for thick photopolymer samples typically packaged in glass. Finally, as shown on the right-hand side of Figure 1, when the object to be measured is immersed in an oil cell and rotated 360°, the transfer function of ODT is both uniform and without cusps or voids (save the typical one at the origin), making it nearly the ideal microscope.

Figure 1. Summary of the metrology options for angle-multiplexed holographic data storage represented in the spatial frequency domain with and without oil immersion. The fine red cross-hatched region at the origin is the index perturbation recorded by the intensity of a 0.6 NA object. The blue linear filled regions are the holograms of this object and their phase conjugates recorded by the 30° reference beam sweep in air. Each line represents a 3° rotation of the reference beam. The dashed black line shows the maximum extents of the transfer function of any transmission microscope (TM). The grey arcs show the measurement points of optical diffraction tomography (ODT) for a planar object in air (left) and full rotation in an oil cell (right). Finally, the coarse green cross-hatched region on the left is the transfer function of a confocal reflection microscope (CRM).
2. DIFFERENTIAL SCANNING TRANSMISSION MICROSCOPY

Given that the major flaw of confocal reflection microscopy is that its passband does not overlap the transmission-like objects of interest, an obvious alternative is transmission confocal microscopy in which the confocal pinhole is moved to the opposite side of the medium. Unfortunately, low spatial frequency index perturbations in the material cause the image of the focus to deflect and blur as the sample is scanned, requiring an adaptive tracking system to maintain the alignment of the pinhole [14]. Kawata, et. al. suggested that the tracking signal for this alignment system in the form of the differential signal from a split detector was in itself a phase-sensitive microscope [15]. This method is phase-sensitive only for grating components that are Bragg-matched by coupled waves that straddle the detector elements, leading to voids in the transfer function passband.

We therefore have investigated [16] a coherently illuminated transmissive scanning microscope that employs a position-sensing detector such as a lateral effect photodiode, rather than an ordinary photodiode or split detector. The extra information provided by the PSD can be used to calculate the complex 3D dielectric distribution – i.e., the volume distribution of both refractive index and absorption – of a weakly diffracting optical sample. The instrument is noteworthy for phase depth sectioning properties that do not depend on a confocal pinhole and for the fact that it’s phase transfer function is relatively flat throughout the region of support, reducing the problem of noise enhancement during reconstruction. Since high frequencies are enhanced by the differentiating transfer function, the achievable resolution may in some cases be higher than that of a non-differentiating microscope of the same numerical aperture.

The optical layout of the instrument is shown in Figure 2. A diode laser at 635 nm is collimated and passed through a polarizing beam splitter (PBS) and a quarter wave plate (QWP) so that the sample is probed with circularly polarized light, which diffracts isotropically. The objective lens is a LightPath 350340 aspheric microscope objective (N.A. = 0.62) which is corrected for a focal depth of 1.2 mm, stopped down to N.A. = 0.3 in order to permit a total scanning depth range of about a millimeter without active spherical aberration correction. Figure 2 also shows a confocal reflection path consisting of a condenser lens, a pinhole, and a conventional photodetector (PD). This illustrates that scanning reflection and transmission microscopy are compatible and, as shown in Figure 1, provide complementary spatial frequency information.

![Optical layout of the instrument](image)

Figure 2. Optical layout of the instrument. The reflection channel on the left implements a CRM, while the position-sensitive detector (PSD) on the right is the detector for the new transmission microscope.

The light transmitted through the sample is collected by a 4-F camera lens relay, and then apertured down to the angular extent of the undeviated probe beam. The relay allows a large working distance on the exit side of the sample despite the small 4.0 mm size of the ON-TRAK PSM2-4 PSD. The PSD detects the x and y centroids of the optical power distribution upon the detector (i.e., beam position), as well as total optical power.
An important item on our requirements list is signal gain to enable sensing the weak objects typical of diffusion photopolymers. In this microscope, the diffracted light mixes coherently with the undepleted probe producing an irradiance modulation that is detected at a specific point on the detector in the far field. This mixing has the effect of providing coherent homodyne gain to the diffracted portion of the light, resulting in significant amplification. This will be demonstrated in the results, below.

A 3D image is collected by scanning the object through a series of sample points where the total optical power, the \( x \) power centroid, and \( y \) power centroid data are recorded. Once complete, the computed transfer function of the instrument is used to post-process the data into volume maps of the absorption and index of refraction. The three independent channels are used in this calculation to produce separate and independent amplitude and phase information, satisfying another of the key requirements.

![Figure 3](image1.png)

**Figure 3** – (a) Contour map of a cross section of the imaginary part of the transfer function mapping the imaginary (absorptive) dielectric to the optical power channel. (b) Corresponding impulse response.

![Figure 4](image2.png)

**Figure 4** – (a) Contour map of a cross section of the imaginary part of the transfer function mapping the real (phase) dielectric to the x PSD channel. (b) Corresponding impulse response.
Images of these transfer functions and their Fourier transforms (impulse responses) are shown in Figures 3 and 4. The amplitude channel of the microscope is just a simple bright-field transmission microscope and thus the transfer function is identical to that shown in Figure 1. Although these calculations assumed a uniformly filled pupil, the transfer function is strongly attenuated at high spatial frequencies, a feature shared by most transmission microscopes. The impulse response, found by Fourier transforming this transfer function, recovers the amplitude profile of that probe beam, which is an Airy disk at the focus. The PSF of the amplitude channel is the magnitude squared of this impulse response. By conservation of energy, the $xy$ integral of the PSF must be independent of depth, $z$, and thus this channel has the poor depth selectivity common to most transmission microscopes.

The transfer function of the $x$ centroid (deflection) channel is shown in Figure 4 and has the same region of support in the frequency domain. However, the differential nature of the detection results in the transfer function being multiplied by $k_x$ in the case of the $x$ centroid channel. This has a number of important consequences. First, the high spatial frequency portions of the transfer function are amplified, resulting in a more uniform passband and higher resolution (note the different contour distribution in Figures 3 and 4). Second, the impulse response (part b) is no longer that of a Gaussian beam and thus is not required to conserve energy in $z$. Thus true depth sectioning is allowed.

We follow the definition of depth selectivity first proposed in [9], namely

\[ psf_{int}(z) \equiv \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} |psf(\rho)| d\rho dy. \]  

(1)

This sectioning function is plotted in Figure 5 for the absorption channel (Figure 3), the phase channel (Figure 4) and a confocal reflection microscope for comparison. This figure shows that the phase channel does reject out-of-focus signals in the same manner as a CRM, although not to the same extent. The ripples versus depth are due to the fully-filled pupil used to simplify the calculation of the transfer functions and thus this depth sectioning can likely be improved by using a smooth probe beam such as a Gaussian.

Figure 5 – Optical Depth sectioning as determined by the integrated magnitude of the impulse response within depth planes. The coordinate $u$ is normalized depth according to [17].
Figure 6 shows sample results for several of the applications of interest. Given the resolution of our demonstration system, the image of the data hologram (left) is mainly the ambiguity term due to the object intensity, which is unknown using just the holographic data channel. Another interesting feature is the line above and below the hologram. This is caused by diffraction off of an aperture earlier in the system [18].

The image on the right is of a 633 nm single mode glass fiber encapsulated into Tapestry polymer. A 3D direct-write lithography system has written a waveguide up to the core of the fiber [8]. The instrument’s sensitivity is demonstrated by the clear contrast between the fiber and the nearly index-matched photopolymer. The 2.2 µm fiber core, which has an index contrast of only $\Delta n = 5 \times 10^{-3}$ vs. the cladding ($1.8 \times 10^{-2}$ waves of delay), is also barely visible. This object has less than $1/50^\text{th}$ total waves of delay, demonstrating the sensitivity of the microscope.

![Figure 6. Demonstration images of the transmission microscope.](image)

These images were taken at a low objective NA of 0.3 in order to limit spherical aberration as the material is moved up to ±500 mm in depth. While this could be overcome with well-known systems that dynamically correct the aberration, it does highlight the inherent difficulty of high-resolution imaging at large working distance. The next system avoids this difficulty.

### 3. OPTICAL DIFFRACTION TOMOGRAPHY

In contrast to traditional microscopy, ODT measures objects directly in the frequency domain. This has a number of advantages, particularly the ability to directly measure deeply-buried, complex multi-dimensional structures with both amplitude and phase features. Like virtually all microscopic methods, quantitative reconstruction is possibly only when the object is weakly scattering. After measurement, an inverse Fourier transform produces the quantitative complex dielectric constant of the object. Below we briefly introduce ODT in its simplest form, then show how it can be extended to measure extremely weak features recorded in volume photopolymer. The reader interested in more detail on ODT is directed to the literature [19].

The basic principles of ODT are shown in Figure 7. The object is illuminated with a plane coherent wave and the first-order scattering is measured. For simplicity, assume this measurement is in the far-field either through use of a Fourier transform lens or propagation. Note in this second case that by using a mechanically scanned detector, the
entire ODT setup can be constructed with no lenses even at large effective numerical aperture, in contrast to typical microscopy where the cost and complexity of the lenses dramatically increase with NA. As shown on the right hand side of Figure 7, the far-field scattering pattern is proportional to the Fourier transform of the object dielectric constant on an arc with radius \( k = 2 \pi n / \lambda \). The object is rotated to reveal different arcs through the Fourier transform, as shown in Figure 1.

If the object is embedded in a planar package, typical of volume photopolymers, total internal reflection limits the range of incident and diffracted angles, restricting the sampled Fourier space to be exactly that of a transmission microscope whose NA equals the maximum tilt angle. However, if the object is immersed in an oil cell with a curved output surface such that it can be rotated arbitrarily and all scattering can escape the cell, the entire Fourier space out to a range \( 2^{1/2} k \) can be measured. Note in Figure 1 that this circular region of support has none of the cusps or narrow tails common to reflection and transmission microscopes. The sampling region of ODT does have a null at the origin since one can not measure the undeflected scattered field that is collocated with the undepleted probe beam. This loss of DC information is common in microscopy and restricts the measurement to the relative dielectric constant, not its absolute level.

In order to experimentally demonstrate an extension to this technique, we have made several simplifications. First, to capture an arbitrary 3D object, the scattering must be captured in 2D to build the full 3D Fourier transform as the object is rotated. As is common in tomography, we will restrict objects to be uniform in the dimension out of the plane, although in principle one can reconstruct a 2D slice, then translate the object through the detection region to sample other planes. Second, since the Fourier transform is generally complex, both the optical amplitude and phase of the scattering must be measured. This can be done via interferometry or by phase-retrieval from intensity measurements at different propagation distances from the object. We will make the simplifying assumption that the Fourier transform is real and positive so that electric field is the square root of intensity. This restricts the method to smooth symmetric objects, but this is sufficient for purposes of demonstration of our extension, described next.

![Figure 7. ODT setup (left) and explanation in Fourier space (right). Scattering from an incident plane wave samples the Fourier transform of the object on a circular arc with radius equal to \( k \) in the material. By rotating the object, a series of curved samples of the Fourier space are recorded. An inverse discrete Fourier transform from the unequally sampled grid shown in Figure 1 produces a real-space image of the object’s complex dielectric constant.](image)

A single micron-scale index feature with index contrast of \( 10^{-4} \) recorded into a photopolymer would produce insufficient diffraction to overcome surface scatter, bulk scatter or detector noise. Since electric field is proportional
to the square root of the measured intensity, capturing four orders of electric field magnitude requires detecting eight orders of intensity range. This demands relatively strong diffraction to keep the full dynamic range at adequate SNR.

For arbitrary objects such as biological specimens one can not increase the diffraction from a fixed object. However, for perturbations written into photopolymer, it is possible to do so. Since these perturbations are written optically, they can be replicated at precise spacing simply by placing the photopolymer on a precision stage. These multiple identical objects are then illuminated with a plane wave such that their diffraction adds coherently at spatial frequencies inverse to the object spacing and destructively elsewhere. The intensity in the diffracted orders is amplified by the square of the number of illuminated objects. This enables virtually arbitrary increase in the diffracted signal level so that extremely weak objects can be imaged.

The concept is illustrated in Figure 8 for index lines ruled into InPhase Tapestry photopolymer by a 1 microwatt Gaussian beam of 0.3 NA moving into the page at 2 mm/second. Since this material is approximately linear, the recorded index change is approximately proportional to the intensity of a Gaussian beam and its Fourier transform on the right of Figure 8 has the characteristic double-leaf region of support shown in previous figures. The dashed lines on the Fourier-space figure show the locations of the diffraction orders. The Fourier transform of the object is thus sampled at these discrete locations and can not be measured elsewhere.

This lost information does not limit the ability of ODT to reconstruct the object, however. Let the largest \( x \) dimension of the object be \( \delta x \) and the spacing between the objects be \( \Delta x \). The smallest feature of the object’s Fourier transform is constrained to be roughly \( \delta k_x \approx 2 \pi / \delta x \). The diffraction orders will be spaced by \( \Delta k_x \approx 2 \pi / \Delta x \). Thus as long as the replicated objects are separated by much more than their width, \( \Delta x >> \delta x \), the sample locations in Fourier space will be finer than the smallest variation in the Fourier transform \( 2 \pi / \Delta x \ll 2 \pi / \delta x \). The Fourier transform of the single object can therefore be accurately characterized by just the discrete samples.

![Figure 8](image_url)  
**Figure 8.** Real space DIC image and calculated Fourier space views of the how replication of the object creates a periodic diffraction pattern. Each grating order is a sample of the Fourier transform of a single object, but amplified in electric field by the number of illuminated objects. Any photopatterned object can be so replicated in order to amplify its diffraction above the noise.

The ODT reconstruction of the objects in Figure 8 is compared to a microtomed slice viewed on a DIC phase-contrast microscope in Figure 9. Note that the noise evident in the DIC image, part a, is not present in the ODT image, part b. This SNR enhancement is due to both greater signal levels at the detector and to averaging any material imperfections over many grating lines.
4. DISCUSSION AND COMPARISON

Figure 10 shows images of a 10 µm gradient index waveguide written into a cm-thick volume of InPhase Tapestry photopolymer. In order to match the transfer functions of the DIC (a) and scanning transmission (b) microscopes, the sample was microtomed and imaged normal to the page. The ODT image (c) was taken directly from an array of waveguides in the cm-thick volume. In this case, since the object is circularly symmetric, a single slice of its Fourier transform is sufficient.

The figure conveniently summarizes our results. The DIC image (a) is somewhat noisy since no amplification is applied. The image is not quantitative and is blurry due to out-of-focus images. These two effects are coupled since reducing the sample thickness will decrease the signal while reducing the out-of-focus material.

The scanning differential transmission image (b) shows much less noise due to the homodyne amplification and has been processed using the theoretical transfer functions to be a quantitative measure of index. The measured profile
is somewhat narrower than (c) and includes small valleys near the index peak. Both are consistent with the fact that the transfer function shown in Figure 4 is very low near the origin, effectively high-pass filtering the object. Further validation is required to fully establish this as the cause, but it illustrates how nonuniform transfer functions are problematic.

Finally, the ODT image is shown in (c). This image shows essentially no noise due to both the diffraction amplification from multiple identical objects and average of material background variation across these multiple samples. The method is nondestructive and has a perfectly flat passband.

In conclusion, we have shown how standard microscopy methods are largely inappropriate for metrology of the deeply buried, weak, high resolution phase and amplitude objects characteristic of optically patterned diffusion polymers. The two instruments described overcome these issues in different ways but both give true 3D, quantitative index measurements. The two methods of signal amplification are shown to enable high SNR images of very weak objects without the traditional tradeoff of resolution and index contrast of thin sections. Metrology such as this is required to fully exploit photopolymer devices and to validate materials models.

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