Bragg holography and holographic polymers

Outline

• Thin holography
• Holographic photopolymers
• Bragg holography
  – Efficiency
  – Selectivity
Basics of holography

Recording the optical phase

- Recording medium must physically respond to intensity (not field)
- We wish to record the complete optical field ("whole drawing")
- Must use interference

**Write**

![Write Diagram]

\[ I_{\text{WRITE}} = |E_O + E_R|^2 \]
\[ = |E_O|^2 + |E_R|^2 + E_O^* E_R + E_O E_R^* \]

**Read**

![Read Diagram]

\[ E_{\text{READ}} = T E_R \]
\[ \propto \left[ |E_O|^2 + |E_R|^2 + E_O^* E_R + E_O E_R^* \right] E_R \]
\[ = \left[ |E_O|^2 + |E_R|^2 \right] E_R + E_O^* E_R E_R + E_O |E_R|^2 \]

- Reconstructed object weighed by intensity of reference
- Must separate terms in angle
Hologram types

A brief glossary

• **Object beam**: the light from the object being recorded
• **Reference beam**: the light that is interfered with the object beam during recording. Also the light used to reconstruct the hologram during playback.
• **Thin vs Thick**: Thin holograms are most common in the commercial world. They exhibit wide angular and spectral replay tolerance. Thick or “Bragg” holograms are used in many research settings. They exhibit narrow angular and/or spectral replay tolerance. This is exploited in holographic data storage.
• **Phase vs amplitude**: Holograms can be made via spatial modulation of the phase or the amplitude of the field. Typically amplitude is modulated via varying absorbance (loss).
• **Phase**: Phase holograms can be formed by spatial modulation of the index of refraction, by spatial modulation of the surface profile or both.
• **Transmission vs reflection**: The object can be reconstructed on the same side from which the reference is incident (reflection) or the opposite (transmission). Security holograms are a common form of reflective, thin, phase, surface-relief hologram.
A perfect (100% to 0% transmission) amplitude (absorption) hologram would have a transmission function like:

$$T \equiv \frac{E_{out}}{E_{in}} = \frac{1}{2} + \frac{1}{2} \cos \left( \frac{2\pi}{\lambda_x} x \right)$$

When illuminated by a reference, the electric field just after the hologram would be:

$$E_{out} = T E_{in} = \frac{E_{in}}{2} + \frac{E_{in}}{4} \left[ \exp \left( j \frac{2\pi}{\lambda_x} x \right) + \exp \left( -j \frac{2\pi}{\lambda_x} x \right) \right]$$

Note that this is expressed as a Fourier transform. Each term corresponds to diffraction into a particular angle, only one of which is the object reconstruction. It’s power efficiency is

$$\eta \equiv \frac{I_{obj-out}}{I_{ref-in}} = \left( \frac{1}{4} \right)^2 = 6.25\%$$
Allocation of bandwidth

Separation of terms

- Let the spatial-frequency bandwidth of the object field $E_O$ be $B_O$
- Let the spatial-frequency bandwidth of the object field $E_R$ be $B_R$
- The bandwidth of $|E_O(x)|^2$ is equal to the bandwidth of $E_O(f_x) \otimes E_O(f_x)$ which is $2B_O$

$$f_O - f_R > \frac{1}{2} \left\{ B_R + \text{Max}[2B_O, 2B_R] + B_O + 2B_R \right\}$$  To separate terms

$$f_O - f_R > \frac{3}{2} B_O$$  Special case for plane-wave reference ($B_R=0$)
Conjugate playback
Time reversal

$Illuminate\ with\ phase\text{-}conjugate\ reference$

$E_{\text{CONJ}} = T E_R^*$

$\propto \left[ |E_O|^2 + |E_R|^2 + E_O^* E_R + E_O E_R^* \right] E_R^*$

$= \left[ |E_O|^2 + |E_R|^2 \right] E_R + \overline{E_O^* E_R^*} + E_O E_R^* E_R^*$

$Use\ in\ imaging\ through\ phase\ (not\ amplitude!)\ perturbation:$

Write

Ref

Object

-phase perturbation

Read

Reconstruction

Ref
Angular bandwidth
Replay with tilted reference beam

\[ \mathcal{E}_{\text{READ}} = T \mathcal{E}_R e^{-j k_x x} \]
\[ \propto \left[ |\mathcal{E}_O|^2 + |\mathcal{E}_R|^2 \right] \mathcal{E}_R e^{-j k_x x} + \mathcal{E}_O^* e^{-j k_x x} \mathcal{E}_R \mathcal{E}_R + \mathcal{E}_O e^{-j k_x x} |\mathcal{E}_R|^2 \]

- Add spatial frequency to reconstruction at plane of hologram.
- Similar to tilt – precisely equivalent to grating deflection.
- Only limitation is TIR = wide angular bandwidth.
Spectral bandwidth
Replay with different color reference beam

\[ \hat{x} \cdot \vec{k}_{R-red} = \hat{x} \cdot \vec{k}_{R-blue} \]

\[ \frac{2\pi}{\lambda_{red}} \sin \theta_{R-red} = \frac{2\pi}{\lambda_{blue}} \sin \theta_{R-blue} \]

\[ n_{eff} \sin \theta_{R-red} = \sin \theta_{R-blue} \]

To reconstruct object at same angle

\[ n_{eff} = \frac{\lambda_{blue}}{\lambda_{red}} \]

- Reconstruction is equivalent to refracting into a material of index \( n_{eff} \)
- Only limitation is available angular bandwidth for ref and obj beams
- Thus, wide spectral bandwidth.
Glossary of polymer chemistry

- **Monomer** “single part”: small building blocks reacted to become...
- **Oligomer** “few parts”: medium-sized molecule of 2 or more monomers
- **Polymer** “many parts”: macromolecule built from many monomers
- **Photolysis** “light cutting”: photoinitiator molecule breaks into two radicals
- **Photoinitiator**: species which absorbs light to start initiation reaction
- **Initiation**: radical reacts with monomer to start propagation reaction
- **Propagation or polymerization**: Chain reaction which adds monomer units
- **Termination**: Reaction which stops propagation, including radical dimerization
- **Inhibitor**: A species such as O$_2$ which reacts with and terminates radicals
- **O$_2$ threshold**: Optical dose required to create sufficient radicals to consume O$_2$
- **Hydrogen abstraction**: Transfer of radical by exchange of a hydrogen nucleus (proton)
- **Chain transfer**: termination of one propagating polymer by release of radical
Typical “two component” chemistry

Initiating radicals attach to the matrix and react with monomer.

“Sponge” model

<table>
<thead>
<tr>
<th>Chemical component</th>
<th>Chemical function</th>
<th>wt %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diphenyle(2,4,6-trimethylbenzoyl)</td>
<td>Photo initiator</td>
<td>0.067</td>
</tr>
<tr>
<td>phosphineoxide (TPO)</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Tribromophenyl Acrylate</strong></td>
<td>Writing monomer</td>
<td>6.00</td>
</tr>
<tr>
<td>Butyl phthalate 99%</td>
<td>Plasticizer</td>
<td>0.50</td>
</tr>
<tr>
<td>Polyester Block Polyether</td>
<td>Matrix component 1</td>
<td>55.61</td>
</tr>
<tr>
<td>Desmodur 3900</td>
<td>Matrix component 2</td>
<td>37.81</td>
</tr>
<tr>
<td>Dibutyltin Dilaurate, 95%</td>
<td>Catalyst</td>
<td>0.01</td>
</tr>
</tbody>
</table>

Photopolymer
- 405 nm initiation
- 1 photon = high speed
- High refractive index
- Small, mobile
- Low shrinkage
- Low scatter

Matrix polymer
- Urethane
- Catalyzed at room T
- Low shrinkage stress
- Phase uniform
- Low scatter
- Low refractive index

Initiating radicals attach to the matrix and react with monomer.

“Sponge” model

- Bragg holography and materials
  - Holographic photopolymers
Photons create mobile radicals, some of which react with matrix to create fixed pattern.
Immobile polymer chains grow from these radicals. Replacement monomer diffuses into reaction region.
**Basic mechanisms**

Matrix swells, decreasing matrix concentration in bright regions.

- Bragg holography and materials
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Basic mechanisms

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Optical flood exposure consumes remaining initiator and monomer.
Basic mechanisms

Segregation of writing polymer and matrix creates index distribution.

- Bragg holography and materials
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Source of refractive index

Mobile writing monomer swells matrix polymer, replacing it 1:1

\[ n = n_{\text{MatrixPolymer}} + n_{\text{WritingPolymer}} \]

\[ = n_{\text{MatrixPolymer}} \left( 1 + n_{\text{WritingPolymer}} \right) + n_{\text{WritingPolymer}} \]

\[ = n_{\text{MatrixPolymer}} + \left( n_{\text{WritingPolymer}} - n_{\text{MatrixPolymer}} \right)\] 

\[ = n_0 + n \]

Validation with confocal Raman spectroscopy:
Typical grating dynamics

\[ \varphi = 0.7 \, \text{m}, \quad I = 100 \, \text{mW cm}^{-2}, \quad t = 3 \, \text{sec} \]

\[ \varphi = 5 \, \text{m}, \quad I = 100 \, \text{mW cm}^{-2}, \quad t = 1 \, \text{sec} \]

\[ \varphi = 0.7 \, \text{m}, \quad I = 10 \, \text{mW cm}^{-2}, \quad t = 3 \, \text{sec} \]

This material is unusually slow. Commercial materials will develop in seconds.
Coupled modes derivation (1/2)

Homogeneous = Fourier optics

Start with the vector wave equation with the material described by a homogeneous and an inhomogeneous permittivity

\[ \nabla \times \nabla \times \vec{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \left[ \bar{\varepsilon}_H + \bar{\varepsilon}_{IH} (\vec{r}) \right] \vec{E} = 0 \]

\[ \nabla \times \nabla \times \vec{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \bar{\varepsilon}_H \vec{E} = - \frac{1}{c^2_0} \frac{\partial^2}{\partial t^2} \bar{\varepsilon}_{IH} (\vec{r}) \vec{E} \]

The solutions to the homogeneous equation (uniform material, RHS=0) are two polarized plane waves. Since these plane waves are complex exponentials in time and space, an incident field on the half-space boundary \( z=0 \) can be expressed as a sum of the eigenfunction plane waves via a Fourier transform in time, \( x \) and \( y \) (here written in the scalar form for simplicity):

\[ \left( \begin{array}{c} k_x, k_y, z = 0 \end{array} \right) = F_{txy} \left[ E(t, x, y, 0) \right] \]

Since these eigenfunctions propagate in \( z \) via accumulation of phase, the fields can be calculated at any distance \( z \) via the transfer function:

\[ E(t, x, y, z) = F_{txy}^{-1} \left\{ \left( \begin{array}{c} k_x, k_y, z = 0 \end{array} \right) e^{jk_z(k_x, k_y)z} \right\} \]

\[ = F_{txy}^{-1} \left\{ F_{txy} \left[ E(t, x, y, 0) \right] e^{jk_z(k_x, k_y)z} \right\} \]

Which is the foundational equation of Fourier optics. Note that the amplitudes of the plane waves are constant with \( z \).
Coupled modes derivation (2/2)

Inhomogeneous

As an approximate solution to the inhomogeneous case, assume that the plane wave amplitudes can exchange energy via coupling and thus vary in $z$.

\[
\left( k_x, k_y, z = 0 \right) = F_{txy} \left[ E(t, x, y, 0) \right]
\]

\[
E(t, x, y, z) = F_{txy}^{-1} \left\{ \left( k_x, k_y, z \right) e^{jk_z \left( k_x, k_y, z \right)} \right\}
\]

… and substitute into the wave-equation. The product of two functions with $z$ dependence will generate multiple terms via the chain rule. All terms on the right hand side which don’t involve $z$-derivatives of the envelope must sum to zero because they are solutions to the homogeneous wave equation, leaving an ordinary DE in $z$ for the envelope which is unfortunately complicated by the RHS:

\[
\frac{d^2 \mathcal{E}}{dz^2} e^{jk_z z} - 2 j k_z \frac{d \mathcal{E}}{dz} e^{jk_z z} \approx -2 j k_z \frac{d \mathcal{E}}{dz} e^{jk_z z} = -k_0^2 F_{txy} \left\{ \mathcal{E}_{IH} \left( \vec{r} \right) E \right\}
\]

where the second derivative has been dropped by the slowly-varying envelope approximation (SVEA). To deal with the RHS, we now assume that, for the problem of interest, only a finite number of modes present. This reduces the continuous Fourier transform integrals to a sum over a discrete number of “modes” = plane waves in this case:

\[
E(t, x, y, z) = \sum_i u_i(z) e^{jk_{z,i} z} = \sum_i \left( t \ k_x \ k_y \right) e^{jk_{z,i} z}
\]

which reduces the equation to a coupled set of first-order ODEs:

\[
\frac{d u_m}{dz} = j k_{z,m} u_m(z) \sum_i j^{m,i} u_i(z)
\]

where $\kappa$ is the coupling between modes caused by the perturbation. Note that if the coupling is zero, the solution is uncoupled plane waves with unchanging amplitude.
Example: Two coupled modes

The equations for the field amplitudes $u_m(z) e^{j m z}$ are

$$\frac{d}{dz} u_0 = j b_0 u_0 - j k u_1$$
$$\frac{d}{dz} u_1 = j b_1 u_1 - j k u_0$$

where $\kappa = \kappa_{1,2} = \kappa_{2,1}$

which have simple solutions:

$$I_0(z) = |u_0(z)|^2 = \frac{\cos^2(g z) + \gamma^2}{1 + \gamma^2}$$
$$I_1(z) = |u_1(z)|^2 = \frac{\sin^2(g z)}{1 + \gamma^2}$$

where $\gamma = \left(\frac{\beta_0 - \beta_1}{\kappa}\right)/2$

Coupling vs. distance
Application to volume holography

If the incident wave is Bragg matched such that $\Delta \beta = 0$,

$$I_{inc}(z) \mid u_{inc}(z) \mid^2 = \cos^2 (z)$$
$$I_{diff}(z) \mid u_{diff}(z) \mid^2 = \sin^2 (z)$$

Thus the efficiency of the hologram (Koglenik solution) is

$$\equiv \frac{I_{diff}(L)}{I_{inc}(0)} = \sin^2 \left( \frac{L \ n}{\cos \ diff \ 0} \right)$$

100% efficiency possible

For weak diffraction, diffracted field grows linearly with $L$ (k-space solution)

$$= \left( \frac{L \ n}{\cos \ diff \ 0} \right)^2$$

If $M$ holograms divide the available $\Delta n$ into equal fractions,

$$M = \left( \frac{L \ n / M}{\cos \ diff \ 0} \right)^2 = \left( \frac{M_#}{M} \right)^2$$

where $M_# = \frac{L \ n}{\cos \ diff \ 0}$

Thus the efficiency of multiplexed holograms falls quadratically with number.
Angular and wavelength selectivity

Consider a hologram of thickness $L$ written by two plane waves, then read out by one of them.

What tilt of the incident wave causes $k_z$ phase mismatch equal to the first null?

$$k_z = 2k \sin$$

$$( k_z ) = \text{Bragg Matched} \ \text{sinc}^2 \left( \frac{k_z L}{2} \right)$$

$$k_{z,Null} = \frac{2}{L} = 2 \ \text{Null} \ k \sin = \frac{2}{\text{Null}}$$

$$\text{Null} = \frac{L}{2}$$

What wavelength shift of the incident wave causes $k_z$ phase mismatch equal to the first null?

$$\frac{2}{L} = (2 \ k \sin) \ \tan$$

$$= \left( \frac{k K}{k} \right) \ \tan$$

$$\frac{k}{k} = \frac{L \tan}{\text{Null}}$$