# CU Holographic Photopolymer modelling code suite

Release 2.2

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## New in this update

* Dynamics of radicals added. There is not an obvious way to validate this because the reaction equations become nonlinear and thus do not admit a harmonic series solution. Thus, this should be treated with some caution.
* Scatter now updated to use Rayleigh scatter. Number density of scatterers is now calculated from requested initial haze. This seems to be slightly inaccurate: initial haze can be off by ~15%. I suspect this is because I assume fully incoherent addition of scatter. However, this is still a significant improvement from the previous approach.
* The code now allows arbitrary reference and object beam angles (for slanted transmission gratings) and read wavelength different than writing.
* Multiplexing is allowed by specification of a vector (one entry for each exposure) of exposure times , incident and diffracted angles.
* Angular Bragg selectivity can now be calculated at multiple arbitrary time points through the variable BraggTSteps
* Ad-hoc phase separation added through variables PSThreshold, PS1 etc. A more rigorous calculation based on Flory Rehner is in preparation so I would recommend not spending much time with the current implementation

## Guide to the functions

Matlab version Developed in Matlab 2020a

### Matlab toolbox functions

fit Matlab function from curve fitting toolbox

divisors Matlab function from symbolic math toolbox

sinc Matlab function from signal processing toolbox

### Functions

CallHoloPolymer The top level routine that calls the numerical, theoretical and rendering routines.

NumericalHoloPolymer Models (3 space + 1 time) dimensional diffraction, scatter, reaction and diffusion of a holographic photopolymer exposed to record one or more transmission gratings. Inputs are specified as a list of name value pairs. All outputs are returned in a single structure. Input and output values are saved by default to “.mat” files for later reuse.

TheoryHoloPolymer Use harmonic expansion of RD equations plus the strong limit theory for index formation to predict holographic photopolymer response vs. exposure time for an unslanted transmission grating. It uses same input and output structures as the numerical function.

RenderHoloPolymer A set of plotting functions to show and compare various results from the numerical and theoretical codes.

ParseInputs This routine accepts, checks and assigns defaults for all input variables.

m0, m1, m2, m3 Closed-form solutions for the first four harmonics of the reaction/diffusion rate equation for monomer.

P0, P1, P2, P3 Closed-form solutions for the first four harmonics of the reaction/diffusion rate equation for polymer.

PolymerRoots Roots of the characteristic equation for the coupled first-order set of differential equations for the harmonics of the monomer.

BPM \_v2 Implements the Fourier Beam Propagation Method to propagate light across the film, refracting and diffracting in response to material structure. Version 2 uses an input parser to streamline calls and is internally cleaned up in preparation for reflection holography.

LorentzLorenz Calculates refractive index of a mixture

Kogelnik\_Transmission Theoretical efficiency of an ideal transmission hologram

Transmission\_BVM “” calculated by beta value method

### From Matlab file exchange

drawcircle Used in rendering results

wavelength2color Used in rendering results

## Input name/value pairs (defined in InputParser)

**Parameter Default Validity Meaning**

### Bragg selectivity parameters

CalcBragg 0 Logical Compute Bragg?

BraggNulls 2 Scalar >0 Angular extent of Bragg readout

NBraggAngles 41 Scalar >0 Number of samples for Bragg readout

### Data files

InSaveFile see IP.m String Input save file name.

OutSaveFile see IP.m String Output save file name.

InLoadFile "" String Load previous save. Default don’t load.

### Material parameters

nmono 1.545 Scalar >0 Monomer index

nP 1.5789 Scalar >0 Polymer index

nM 1.476 Scalar >0 Matrix index

sigma 0.14 Scalar >0 Full conv shrinkage

phim0 0.1884 Scalar >0 Monomer volume fraction at 20 wt%

mu 0 Scalar >0 Attenuation coefficient[1/m]

Z 10 micro Scalar >0 Sample thickness

Rm 0.005 Scalar >0 D\_monomer K^2 / R\_P

Rr 0 Scalar >0 D\_radical K^2 / R\_A

tau 2.5 Scalar >0 Exposure time multiplied by MEAN

polymerization rate

### Optical parameters

lambda0Wrt 405 nano Scalar >0 Optical wavelength for writing

lambda0Red 405 nano Scalar >0 Optical wavelength for reading

ThetaIncWrt 0.1346 Scalar Internal reference write angle, rad

ThetaDifWrt -0.1346 Scalar Internal object write angle, rad

BeamRatio 1 Scalar >0 Object to reference power ratio

ThetaIncRed Bragg match Scalar Internal reference read angle, rad

w0wrt 100 micro Scalar Waist radius of writing beams

w0red 20 micro Scalar Waist radius of reading beams

### Grid parameters

Nt 20 Scalar >0 Number of simulation time steps

Nx 1024 Even Number of x cells. Best if 2^N

Ny 1024 Even Number of y cells. Best if 2^N

Nz 36 Even Number of z cells

Dx 203 nano Scalar x cell size (Nyquist in air)

Dy 203 nano Scalar y cell size (Nyquist in air)

### Scatter parameters

InitialHaze 0.01 Scalar >0 Haze at t=0 due to scatter

NScatInVol 36000 Scalar >0 N scatter centers in polymer

NScatInSub 0 Scalar >0 N scatter centers in 1st substrate

ScatType ‘none’ ScatType 'RayleighV', 'RayleighS', 'Isotropic',

'None'. Angular spectrum of scatter

NHaxeVsZ 1 Scalar >0 Number of z slices to measure haze

### Phase separation parameters (not recommended – better theory now available)

PSThreshold -1 Scalar Threshold polymer value at onset of

phase sep. Norm to [m0].

Set <0 to turn off

PS1 0.1 Scalar Linear term in dudp polynomial

PS2 0.1 Scalar Quadratic term in dudp polynomial

PS3 0.1 Scalar Third-order term in dudp polynomial

### Distortion parameters

PostDisplace 0 Scalar Apply displacement after recording?

MaxdVOverV -0.026 Scalar Peak volume expansion coeficient at

Gaussian center

## A note on the time index

When a certain number of time steps Nt are requested, the code will return Nt+3 time indices. The meaning of the time index is:

Index Condition

1 Initial condition before exposure

2:Nt+1 Nt steps of the exposure

Nt+2 Dark development after exposure via full diffusion of monomer

Nt+3 Flood cure via complete conversion of monomer

## Output variables (all in structure “out”)

**Parameter Meaning**

### Version tracking

Version Release version number

VersionDate Release version date

### Miscellaneous calculations and defaults

HazeToStop Half angle, in degrees, of haze stop. Set to 2.5 degrees.

thetaB Bragg angle in radians. 0.1364 by default.

n0 Index of material before exposure. With defaults, n0 = 1.4887

nprime Imaginary part of n0. Zero by default

### Coordinates

X Transverse grid size in X in meters. 207.87e-06 by default

Y Transverse grid size in Y in meters. 207.87e-06 by default

dz Step size in z. Quite small for scatter dynamics. 2.7778e-07 by default

x\_x Vector of x coordinates versus x. 1024 elements by default.

y\_y Vector of y coordinates versus y. 1024 elements by default.

z\_z Vector of z coordinates versus z. 36 elements by default.

fx\_fx Vector of fx coordinates versus fx. 1024 elements by default.

fy\_fy Vector of fy coordinates versus fy. 1024 elements by default.

### Haze and scatter

ScatDensity Scatter centers per m^3

dnS\_xyz Index contrast of initial scattering sources. 1024×1024×36 by default

Haze Haze versus time.

HazeX Haze in X slice of width HazeToStop versus time.

Includes Bragg plane so includes mismatched diffraction

HazeY Haze in Y slice of width HazeToStop versus time.

### Index contrast and diffraction efficiency

dn\_xyz Final index contrast. 1024×1024×36 by default

Eta Diffraction efficiency versus time. Nt+3 = 23 elements by default.

DnNum Largest peak to mean index change via numerical fit versus time.

### Harmonics of monomer and polymer

m0Num Numerically fit 0th harmonic of the monomer versus time.

m1Num Numerically fit 1st harmonic of the monomer versus time.

P0Num Numerically fit 0th harmonic of the polymer versus time.

P1Num Numerically fit 1st harmonic of the polymer versus time.

P2Num Numerically fit 2nd harmonic of the polymer versus time.

P3Num Numerically fit 3rd harmonic of the polymer versus time.

### Bragg angular selectivity

HoloDTheta\_theta Bragg angular readout angles. 41 elements by default.

Eta\_theta Bragg efficiency versus angular detuning. 41 elements by default.

### Electric field slices

Eref\_xyZ E field exiting material (z=Z) during Bragg readout. 1024×1024 by default.

Eref\_x0 E field slice (y=0) during Bragg readout. 1024×36 by default.

Enrm\_xyZ E field exiting material (z=Z) during normal incident (haze) readout.

1024×1024 by default.

### Added by TheoryHoloPolymer

m0Thy Theoretically calculated 0th harmonic of the monomer versus time.

m1Thy Theoretically calculated 1st harmonic of the monomer versus time.

P0Thy Theoretically calculated 0th harmonic of the polymer versus time.

P1Thy Theoretically calculated 1st harmonic of the polymer versus time.

P2Thy Theoretically calculated 2nd harmonic of the polymer versus time.

P3Thy Theoretically calculated 3rd harmonic of the polymer versus time.

DnThy Theoretically calculated index contrast as a function of time.

DnThyMax Largest possible index contrast calculated by theory. 0.0162 by default.

# Reaction diffusion equations

The first-order rate equation for radical diffusion, first-order termination, and generation is

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where is the diffusivity of radicals, the unimolecular termination rate, is initiation rate incorporating the initiator details, is the spatial average intensity with units of W/cm2 and is the hologram period. Radicals react with monomer as described by the first-order propagation rate equation

where is monomer diffusivity and is the second-order rate constant of propagation. Oligomers are not tracked. Polymer concentration is measured with units of monomer and is assumed to be immobile upon reaction such that

Polymer is thus found from the time integral of this equation.

**Normalization**

Define the rate of initiation (radical generation)

By assuming radicals generation and termination are balanced, we find that the steady state radical concentration in the absence of diffusion is . Normalizing time to the termination rate , space to the hologram period , radical concentration to the steady-state concentration we may write the initiation rate equation as

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where the radical diffusion rate to reaction rate constant is

This unitless ratio controls the radical distribution. When initiation is reaction limited, , the diffusion rate is much faster than the termination reaction and the radical distribution will be largely uniform in space. This occurs at high spatial frequencies and is responsible for index contrast falling proportional to the square of spatial frequency. Conversely, when , initiation is diffusion limited and the radical spatial distribution will be a faithful copy of the intensity pattern. The code supports two approaches to modelling the initiation reaction, controlled by the value of . If is left at the default value of zero, radicals are found from the steady-state initiation equation. No blurring of radicals due to diffusion occurs. Otherwise, radical dynamics are modelled via the initiation rate equation controlled by the parameter .

Similarly, the propagation rate equation can be normalized by first noting that the spatially-averaged propagation rate is . This may be used to normalized time as . Monomer and polymer concentration are normalized to the formulated monomer concentration to yield the normalized propagation rate equation

Analogous to the previous discussion, is the unitless diffusion rate to average consumption rate of monomer. When monomer is reaction limited, , the monomer distribution stays approximately spatially uniform during exposure, suppressing higher harmonics caused by depletion of monomer at intensity maxima. Conversely, the diffusion limited regime results in low fidelity recording of strong holograms that consume a significant fraction of the monomer.

In summary, material dynamics is controlled by three parameters. The first is the exposure duration, given in normalized units The dynamics of radicals and monomer are controlled by just two unitless parameters, and . Ideal recording occurs when the former is zero and the latter is infinity. In this limit, monomer decreases exponentially in time and stays spatially uniform. Hologram growth, as measured by the zero and first harmonic of the normalized polymer distribution, is simply . This shows how the exposure time naturally allows one to simulate weak () and strong ( hologram recording without use of initiator and intensity details. When is finite, the monomer concentration decreases at the intensity maxima, resulting in non-ideal recording and the growth of undesired harmonics, as shown below. Higher harmonics decrease the growth rate of the desired first-order harmonic, reducing the measured dynamic range of the material.

