Polarization properties of gelatin holograms

Dichromated gelatin (DCG) exhibits variable changes in effective refractive index (n) from 1.54 before exposure to less than 1.25 as it expands during processing. This aerogel like effect causes aberrations in diffractive optics and Kogelnik's theory predicts strong polarization separation in gratings at many different angles other than 90 degrees. The diffraction efficiency of both S and P polarizations at any angle is dependent on the product of thickness and index modulation while the angle inside the medium is dependent on n. We investigated predicted conditions where only one polarization would be diffracted and subsequently proved n varies from about 1.4 to 1.2 after processing and depends on the film thickness and processing procedures. Transmission gratings made at angles from 36 to 66 degrees were fit to mathematical models as proof of the phenomena, some performed with extinction ratios greater than 100:1. We were also able to demonstrate a similar range in conformal reflection structures and to design a novel polarizer. The calculation of exposure geometries for display holograms becomes more accurate when index change is included in the formulas but some results remain hard to explain.

REDISCOVERY OF A LOW INDEX

Several years ago we had verified by modeling notch filters that Shankoff was correct in claiming that dichromated gelatin could have an index modulation, delta n, of as much as .26. In 1977 Meyerhofer argued that this may imply that the bulk index, n, could drop as low as 1.27 and that the large delta n and corresponding low n was the result of forming small distributed voids of air in the gelatin. He offered no proof or experimental evidence of the low value and made no comments pertaining to the implications and as far as we know it has been overlooked and undervalued ever since then.

We were made aware of Meyerhofer's conclusions and publication, which was on my bookshelf for 10 years, only after we had tabulated hundreds of data points that appeared to support values of n as low as 1.2. We were attempting to make a polarization splitter at an angle that should have worked if the n were near 1.5, in desperation we tried incremental angles above and below the calculated angle. We made hundreds of gratings and carefully plotted the S and P efficiencies as a function of delta n in the region where the S first goes to zero. The design we were concentrating on depended on an internal diffraction angle of 30 degrees, which of course depended strongly on n for a grating structure with fringes normal to the surface. The experimental data we gathered is shown graphically in figure 1.

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Figure 1. Efficiency of S and P as a function of delta n for angles between 36 and 50 degrees, experimental results in 7 micron films, P curve sections are drawn over a common minimum S value.

Transmission proofs of a low effective n

The data we had in hand indicated by modeling on a Mathcad template that the index was around 1.26 and we eventually modeled 4 other configurations that could be used to demonstrate a low n or measure it more closely. Each model was derived from Kogelnik's one dimensional coupled wave model of either transmission or reflection gratings. Figure 2 is the model that best matched to the first experimental data in figure 1, the thin lines are computed plots and the heavy sections indicate where supporting data was obtained by making and testing at 38 degrees until both crossing points were found. The crossing points had to be nearly equal for further evidence of being at the right angle. Figure 3 is a simple proof where the internal angle will be 90 degrees and no P can be diffracted at any value of delta n. This proof has a catch to it because the value of n varies with the value of delta n so some p light actually will be diffracted at low values of delta n where n is still high. The range of delta n we tested pushed n low enough to nearly zero out the P light as S went through 2 maxima at 64 degrees external. Figure 4 is an accidental proof that we modeled after the fact as we reached modulation saturation while testing at 48 degrees. At an external angle of 48 degrees we made a few gratings that diffracted virtually all of both polarizations which models to an n of about 1.27. Some of the data points in figures 2 thru 4 were taken from 7 micron films and also in 5 micron films which because of gradients behave
more like 4 micron films. The positions of maxima and minima remained the same for both films.

Figure 2. Computed model with overlay of normalized experimental data points taken from 35 samples.

Figure 3. Computed model for 90 degree turn internally with some data points normalized and overlaid.
Figure 4. Computed model matching experimental results of S and P both near 100% in 2 or 3 samples.

Reflection proofs of a low n in DCG

Figure 5 shows what turned out to be the most conclusive and easiest to demonstrate proof. Conformal reflectors have an angular spectral bandwidth that varies widely with n. The first plot shows this dispersion curve for n = 1.54 and a zone where we actually measured various reflectors that had values of n ranging from 1.4 to 1.27. The small straight horizontal line is the locus of external incident angles where no P light is reflected and runs from 64 to about 85 degrees.
Figure 5. Computed angular dispersion curves for conformal reflectors with bulk n ranging from 1.54 to 1.27 overlaid with measured data.

Reflection: Internal angles remain constant

Figure 6. Graphical representation of the behavior of a reflector if original exposure conditions prevailed and below it the behavior due to low n and expansion of the film.

Figure 6 illustrates graphically what occurs at an n of 1.27 and what would have to occur at any n above 1.41 where total internal reflection would result whenever internal angles reached 90 degrees. A reflector that worked at 650 nm at normal incidence was made and tested with a laser source at 458 nm for reflected components around the predicted input angle of 64 degrees and the minimum P was found near 64 while S was almost all reflected. Other samples were made and tried in a similar fashion and we found that broadband processed thin films fit the lower dispersion curve and thick narrow band films fit the curve corresponding to an n of about 1.4. The dispersion was particularly easy to measure and this final proof also led to some novel flat plate polarizer designs.

Evidence of a gradient in n
One of our most used film formulae produces an unexposed thickness of 5 microns, an exposed and processed thickness of 5 microns and a processed but collapsed thickness of 3.5 microns. The mixture is 25% sensitizer, most of which is removed during processing, and these measurements make sense but the diffraction properties are a little difficult to explain. A conformal reflector recorded at 514 nm and normal incidence reflects at a center lambda of 580 nm and a bandwidth of 50 nm. It has a weak reflection band extending to about 450 nm and by measuring the dispersion and polarizer angle we know it has an average n of 1.4. If it were uniform and if air replaced the dissolved solids it would have to measure 1.23 times thicker, (over 6 microns), than it actually is to reflect yellow light. It also would not reflect blue light unless some portion of it actually became thinner by about the same amount.

This observation leads us to propose that the microstructure actually has a region of net shrinkage with a corresponding high n and low delta n near its glass interface and a region of net expansion of up to 1.5 times and a corresponding low n and very high delta n. From prior work in modeling chirped and graded reflectors we think this is a reasonable conclusion, the only new observation is a probable and reasonable gradient in n. The before and after structures are shown in figure 7.

**Bragg plane spacing before and after processing**

**Figure 7.** A graphical view of the probable micro structure of a 5 micron thick chirped DCG reflector based on its optical properties.

**IMPLICATIONS OF A LOW N IN DCG DEVICES**

The list of problems and advantages of the low n is long and curious. The first advantage we noted was that the original polarization splitters we were working on could now function at 36 or 38 degrees instead of the 53 degrees we originally calculated to be necessary. Fresnel reflections are lower and cross sections are higher at 36 than they are at 53 and everyone is happier. Some other consequences, advantages and design considerations are enumerated below.
Holographic scanners

The design of “holograms” in DCG sometimes includes consideration of efficiencies in both P and S polarizations. In general as the angles get larger the combined efficiencies go down, except for a few special angles where delta n * T products can catch S and P both at their peaks if carefully controlled. The designs are usually made with an assumed n of 1.5 to 1.56 and measured efficiencies are usually lower than expected because the final n is really less than 1.4. The full internal angles have become larger than the original exposure angles driving P downward. In designs with slanted fringes, the Bragg condition can rarely be satisfied at the construction input angles after processing. The Bragg error appears to come from a net change in film thickness which tilts the fringes up a little so new compensating exposure angles are calculated to compensate. This correction inadvertently also corrects for the change in n which manifests itself exactly the same as a standing up fringe, closer examination would show that the processed film may have expanded by 10% or less but the correction that was finally necessary was calculated for an apparent expansion of 30%. The excess correction is for the lower value of n.

Figure 8 illustrates the effects of expansion alone and then for combined expansion and decreased n in a slanted grating at 633nm. The lower n alters the effective spatial frequency in tilted structures by making the optical path shorter and causing diffraction at a larger full internal angle. The correction for n is essentially an over correction for tilt. In the example the final fringe tilt will be 5 degrees higher than the tilt required in an unchanging n material. Corrections are made and shown for exposure at 488nm.
Figure 8. Highly slanted grating with corrections for a shorter wavelength, expanded film and lowered n.

**Holographic zone plates**

An optic with a varying spatial frequency and a varying fringe tilt angle will reconstruct with aberrations if the n changes between construction and reconstruction. An on axis zone plate producing an f/2 cone of light will have an effective increase of spatial frequency in the radial direction so that at the outer edges the diffraction angle could be off as much as .5 mrad. The focal length gets shorter the further off axis the light gets for zone plates requiring collimated inputs and focusing outputs. This is equivalent to a plano-convex lens except that the spherical aberration is in proportion to the net change in n. This implies that if the exposure is made with a plano convex lens instead of a pinhole then the spot size on reconstruction will more closely meet diffraction limited dimensions. Very low f# optics will have problems closer to those in figure 8 and are very difficult to make with precision and correct fringe tilt.

If the zone plate is designed without slanted fringes, the equivalent of a double convex lens, then it will not be affected by a change in n. This is in contrast to the refractive equivalent which has considerable spherical aberration. Fringes formed without tilt always diffract at the same external angles no matter what n changes to. Multiple exposure gratings with small tilts and rotations have to be designed with a consideration for a change in n even if no compensation is made for
expansion. The change in n usually dominates as a cause for playback errors in any transmission optic that has a tilt in the fringes.

**Multicolor reflection holograms**

A two color reflection hologram can be made with two color exposures and if n and T remain constant then playback will be at the same angles and colors. A good DCG film mixture for two color holograms is 12 microns thick before exposure and 14 to 15 microns thick after exposure and has an initial 6% solid sensitizer concentration. The film expands by a factor of 1.3 causing n to drop to 1.4, resulting in reconstruction at longer wavelengths and different angles. An example of a 2 color design is given to illustrate the relative magnitudes of the changes and the corrections to compensate for them.

If we wish to reconstruct at 560nm and 620 nm with a white light incident at 45 degrees, then we must expose at 60 degrees with 458nm and at 51 degrees with 514nm laser light. We should change the input recording angle by 9 degrees between exposures to correct for the change in n and T. In the reflection geometry the bulk of the correction is for the expansion rather than the drop in n. The reflective geometry magnifies fringe tilt errors more than the transmission geometry. The lower n result in an increase in the angular dispersion so that in general DCG reflection holograms change color over a wider range with equal tilt than equivalent silver grain reflection holograms which have a higher average n.

**Powered reflectors**

We have produced near on axis reflectors of about f/7 that were constructed and reconstructed at 488 but had enormous aberrations. The focal length was observed to vary by over 2% from center to edge of these reflectors. We always had reasoned that if the reconstruction wavelength were identical to the construction wavelength then the aberrations had to be present in the construction optics if they were observed in the reconstruction. This is clearly not the case for any structure with tilted fringes and a changing n. We had assumed that because the reconstruction was at the same wavelength that no fringes could be distorted because the fringe placement must be identical to the construction geometry.

The correct reconstruction color is the result of the film expanding while the index is dropping. The effect of tilted fringes for the reflection case is similar to the transmission case. The effective playback spatial frequency is decreased, the fringes are tilted up a little, the internal angles are the same but the lower n decreases the exit angle giving the optic an increasingly longer focal length as light moves radially outward. Since this is a variable effect the correction can only be iterative and again may be approached by using oppositely aberrated construction waves rather than near perfect spherical waves from pinholes. Construction waves of different curvature may also be used to correct the aberrations in the same fashion that is done to correct for a wavelength shift from construction to reconstruction.

**Reflective polarizers and notch filters**
Notch filters of high density and low n exhibit angular dispersions that span the visible spectrum. One size fits all, we have examples that reflect 650nm at normal incidence and 400 nm at 85 degrees. In fact as mentioned earlier, this dispersion is one of the most convincing proofs of a low average n in these structures. The range of useful polarization separation angles actually runs from about Brewster’s angle to 85 degrees in a simple conformal geometry as shown in figure 9. This design is easy to build and easy to make broadband through processing tricks and multiple exposures. Conformal reflectors are the unique case where the diffraction angle for all colors can be the same and the angle for zero reflection of P light is also the same at all reflected wavelengths.

A second more complex and untested design is shown in figure 10. It will have some of the properties of a transmission hologram and will not have a very broad bandwidth and will be very difficult to make correctly. The only reason for even doing so is that it may be used close to on axis in a flatter package for illumination of LCD panels and other devices requiring polarized light.

White Light Polarizer

**Figure 9.** A simple to make white light polarizer or polarization separator with proven performance.
**Polarizer With Tilted Fringes**

**Figure 10.** A much more difficult to make polarizer with somewhat unknown and untested properties but near on axis operation. The practical angles and bandwidths are unknown.

**EVALUATION AND CONCLUSIONS**

We have illustrated by example and mathematical modelling that DCG always reconstructs with a lower $n$ than it has at the time of exposure. We call this an aerogel effect because it is accomplished with similar procedures to those used in making glass aerogels and exhibits similar properties. Unexposed processed plates of gelatin also expand by as much as 50% and must also exhibit properties of a lower $n$. Wetted and dried processed plates revert to a higher $n$ and corresponding smaller thickness.

A curious side issue is that researchers measuring $n$ with Brewster angle methods or index matching methods do not get the low $n$ values that we have observed and demonstrated. These methods measure $n$ at 1 surface only and as we have observed the $n$ may vary widely through the volume. At least one group at Kaiser Optical has published bulk $n$ measurements as low as 1.34 in the past year and others are studying the gradients in $n$. We have measured the average "effective" bulk $n$ in periodic structures and found that it is important to the design of precision DCG optics of all kinds. Several diffuse object holograms were also tested for a low $n$ and they also exhibit the polarization sensitivity and angular dispersion found in notch filters. We have included these effects in our design software and have found that the model more closely resembles the product, an inclusion of the gradient in the value of $n$ and a curve in the fringes in our modelling would make it even more precise. We will likely pursue these improvements in the future in order to further reduce the surprises we often get.

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