Direct stress optic coefficients for YTZP ceramic and PTFE at GHz frequencies

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Abstract: We report the first measurement of the direct stress optic coefficient for yttria-partially stabilized zirconia (YTZP) ceramic, using illumination between 260 and 380 GHz with applied stresses up to 27 MPa. YTZP exhibited a linear change in refractive index as a function of stress across the entire applied stress domain. A direct stress optic coefficient was also measured for polytetrafluoroethylene (PTFE). PTFE showed viscoelastic behavior at stress values above 4.5 MPa. These results open the way for quantitative sub-surface stress measurements in structural ceramics and ceramic coating systems at GHz and THz frequencies.

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References and links

1. Introduction

Photoelasticity is an experimental technique used to measure sub-surface stress distributions in materials that exhibit stress induced birefringence [1,2]. However, test materials must be transparent at the wavelength used. At optical wavelengths, typical materials include glass and epoxy resins [3,4]. Two options currently exist in order to study stress distributions of non-transparent materials using photoelasticity. The first is to substitute an actual component with a model made from a transparent photoelastic material [1]. Alternatively, a transparent photoelastic coating [5] may be applied to the surface of the actual component.

The purpose of this paper is to determine if ceramics, specifically yttria-partially stabilized zirconia (YTZP), exhibit stress induced birefringence when illuminated with GHz radiation. The approach taken is to measure any change in the ceramic specimen’s refractive index as the applied stress is changed and hence determine the stress optic coefficient. Observation of stress induced birefringence in ceramics has not been reported previously: its presence would demonstrate the feasibility of quantitative sub-surface stress measurements in structural ceramics and ceramic coating systems. Many other materials are transparent at GHz and THz frequencies [6], and the measurement of a stress optic coefficient would enable photoelasticity for visibly opaque materials in which the actual component is tested instead of a model made from a photoelastic material [1]. Alternatively, a transparent photoelastic coating [5] may be applied to the surface of the actual component.

Materials that show stress induced birefringence can be characterized by their stress optic coefficient. Consider the plane stress in a material, at a point, described by principal stresses \( \sigma_1 \) and \( \sigma_2 \). Then,

\[
\begin{align*}
  n_1 - n &= c_1 \sigma_1 - c_2 \sigma_2 \\
  n_2 - n &= c_1 \sigma_2 - c_2 \sigma_1
\end{align*}
\]  

(1)

where \( n \) is the refractive index of the unloaded material and \( n_1 \) and \( n_2 \) are the refractive indices in the direction of the principal stresses \( \sigma_1 \) and \( \sigma_2 \). The stress optic coefficient is defined to be \( C = c_1 + c_2 \), where \( c_1 \) is the direct stress optic coefficient and \( c_2 \) is the transverse stress optic coefficient [1]. In general,

\[
(n_1 - n_2) = C(\sigma_1 - \sigma_2)
\]  

(2)

The unloaded refractive index \( n \) is not needed when measurements are taken at successive stress values. In such a case, if a uniaxial load is applied such that the transverse stress \( \sigma_2 \) is approximately zero, Eq. (1) becomes,

\[
\Delta n_1 = c_1 \Delta \sigma_1
\]  

(3)

Quantitative photoelasticity is based on the linear relationship between changes in refractive index and the applied stress, although photo-viscoelastic behavior in some materials at higher stresses is observed [11–13]. Within the linear elastic regime, changes in the refractive index can be related to changes in strain [14], if the tensile modulus of the material is known.

Intrinsic birefringence of materials at GHz and THz frequencies is routinely measured with techniques such as time-domain spectroscopy (TDS) [11,15]. However, the emphasis of
this paper is the measurement of stress induced (or artificial) birefringence. Conventional THz-TDS has also been used to image through ceramics, in particular thermal barrier coatings [7,8]. In that work, the reflected signals from the interface of YTZP ceramic coatings on a metal substrate were compared between their initial and aged condition after approximately 2000 hours in a furnace. Increased delays in the arrival times of terahertz pulses, as well as changes in the width and shape of the pulses, were observed and attributed to the growth of a thermally grown oxide layer and development of air gaps that would ultimately lead to spalling of the coating. However, these relative changes were not related to any physical property of the ceramic, such as its refractive index.

Ebara et al. [11] reported in a single paragraph of a conference paper the use of conventional THz-TDS with 0.5-1.0 THz illumination to obtain a value of the (direct) stress optic coefficient for PTFE of $c_1 = -14.1 \text{cm}^2 / \text{kgf} = -143.9 / \text{MPa}$ for loads up to ~0.6 MPa. Beyond a load of ~1 MPa, the birefringence increased nonlinearly and did not return to the same birefringence state after the stress was removed. In two linked papers, Song et al. [16], and Li et al. [17], used THz-TDS with 0.3-2.0 THz illumination to measure the (direct) stress optic coefficient without (+ 5.15 /GPa) and with (+ 8.40 /GPa) changes in the specimen thickness considered, respectively.

In this paper, we report the first observation of stress induced birefringence in a ceramic, specifically yttria-partially stabilized zirconia (YTZP), by measuring the direct stress-optic coefficient. Changes in refractive index of ceramic specimens were measured as a function of applied stress, from which the direct stress optic coefficient was determined. A direct stress optic coefficient for polytetrafluoroethylene (PTFE) was also measured in order to investigate the discrepancy between previously published values. The measurements reported in this paper are made with 260-380 GHz illumination.

2. Experimental set-up

Figure 1 shows a schematic of the experimental set-up, a plane polariscope comprising a source, polariser, specimen, analyser and detector. The source (S) was a Virginia Diodes synthesizer and amplifier/multiplier chain that generated a continuous-wave signal tunable between 260 and 380 GHz. A diagonal feed horn launched a vertically polarised, diverging electromagnetic wave that was focused onto the sample, with a spot size of approximately 5 mm, using a pair of 50 mm diameter PTFE plano-convex lenses (L). A linear polariser (P) was placed between the sample and focusing lens with its transmission axis parallel to the plane polarised source, i.e. vertical. A second polariser acting as an analyser (A) was placed after the specimen, also with its transmission axis vertical, i.e. bight field polariscope configuration. A second pair of PTFE plano-convex lenses coupled the beam to a pyroelectric detector (D) (Gentec eo THZ9D). All the measurements were conducted in an anechoic chamber comprising RF absorbing tiles. Additional RF absorbing titles were placed around the aperture of the feed horn and the detector. Both the polarizer and analyser were tilted to an incidence angle of 45° with respect to the optical axis, as shown in Fig. 1, to reduce standing waves between the sample and focusing lenses.

![Fig. 1. Schematic of the experimental arrangement used to measure the stress optic coefficient. Source (S), detector (D), plano-convex lens (L), polarizer (P) and analyser (A).](image-url)
A Deben 2kN dual leadscrew tensile testing stage was used to load the samples. Figure 2(a) shows the tensile stage, with a sample, in the polariscope. Load was applied parallel to the polarisation axis of the illumination. Figure 2(b) shows the geometry of the YTZP and PTFE specimens tested.

Fig. 2. Specimen used for tensile tests. (a) Specimen in Deben tensile stage with polariser and analyser visible, tilted with respect to the optical axis to reduce standing wave interference. (b) Specimen dimensions, in millimeters, with inner dowel pin holes 3.1 mm diameter and outer M4 clearance holes 4.2 mm in diameter.

Three YTZP ceramic (Technox® 2000) samples were sintered and fired at 5 mm thickness by CoorsTek [18]. Each specimen was then ground down to a thickness of 656 ± 5 μm in order to obtain sufficient transmission through the polariscope. Calculation of the initial sample thickness is described in the next section. The grinding direction was parallel to the loading axis of the samples, and therefore parallel to the polarisation axis of the illumination. The YTZP had a density of 6.02 g/cm³, an average crystal size of less than one micron, a Young’s modulus of 210 GPa and Poisson’s ratio of 0.3, values supplied by the manufacturer.

PTFE samples were laser cut from extruded sheet and had an average thickness of 2.177 ± 0.034 mm. Again, calculation of the initial sample thickness is described in the next section. One set of six samples were cut with the loading axis perpendicular to the direction of extrusion from the bulk PTFE sheet. A second set of six samples were cut with the loading axis parallel to the direction of extrusion from the same PTFE sheet. The PTFE had a Young’s modulus of 500 MPa and Poisson’s ratio of 0.46, values supplied by the manufacturer. We confirmed the Young’s modulus value in an independent experiment.

3. Measurement procedure

Before loading each sample, the tensile stage load cell was balanced, because clamping samples into the stage inevitably creates a small applied load. Balancing simply involves setting the tensile stage to apply a load of 0 N once the specimen has been clamped in place. In this manner, the absolute load applied to each sample could be measured. For each specimen, fifteen refractive index measurements were taken at constant load settings between 0 and 350 N, with a 25 N spacing. Sample extension was adjusted by the tensile stage in order to maintain a constant load during each refractive index measurement. For YTZP at all loads, and PTFE at lower loads, any change in extension was minimal during the
measurement period, as shown in Fig. 3(a). Note that the maximum extension in the figure has been normalized for comparison, and is approximately 30 times smaller for YTZP than for PTFE at the same load. Creep became apparent at higher loads for PTFE samples. The initial and final elongation of the sample was recorded during measurement of the refractive index in order to show the associated uncertainty in the applied stress. An appreciable change in the strain due to creep is associated with the onset of photo-viscoelastic effects, which are not the subject of this paper.

At each constant load applied to a specimen, the frequency of the illumination was scanned between 260 and 380 GHz in steps of 0.25 GHz and the signal transmitted by the specimen was recorded. A frequency scan without the specimen was also recorded so that frequency related variations in source emission and detector response could be normalized. A typical normalized spectrum for YTZP is shown in Fig. 3(b). Increased absorption by the YTZP specimen as the frequency increases is shown by a reduction in peak height.

The complex refractive index was extracted from each normalized frequency spectrum, using a nonlinear least squares fit of the Fresnel equations for a single dielectric slab. If the dielectric is surrounded by air ($\n_{air} = 1$), the transmission coefficient is [19,20],

$$T = \frac{\tau_1 \tau_2 e^{-2i\kappa l}}{1 + \rho_1 \rho_2 e^{-2i\kappa l}}$$  \hspace{1cm} (4)

where $\rho_1$, $\rho_2$, $\tau_1$ and $\tau_2$ are the Fresnel coefficients,

$$\rho_1 = \frac{1 - n}{1 + n}$$

$$\rho_2 = \frac{n - 1}{n + 1}$$

$$\tau_1 = \rho_1 + 1$$

$$\tau_2 = \rho_2 + 1$$  \hspace{1cm} (5)

where $n$ is the complex material refractive index, $\kappa$ is the propagation wave number and $l$ is the sample thickness. For a dielectric with appreciable absorption, such as YTZP, $n$ and hence $T$ are represented as complex quantities. The real part of $n$ is the refractive index, $n$, and the imaginary part is $(\alpha / 2 \omega)$, where $\alpha$ is the absorption coefficient, $c$ is the speed of light in a vacuum and $\omega$ is the angular frequency of the illumination. The power measured by the
detector is $T$ multiplied by its complex conjugate, resulting in a real valued function to which the experimentally measured transmitted power was fitted.

Initially, each sample thickness was measured using a digital micrometer with a 1 µm resolution. Using this thickness as an initial estimate, a non-linear least squares fit of the Fresnel equation was performed for each sample for the measured 0 N frequency spectrum, in which the thickness, refractive index and absorption coefficient were allowed to vary. For subsequent measurements at increased constant loads, this initial measured thickness was held constant whilst the refractive index and absorption coefficient were allowed to vary. Figure 3(b) shows an example of the fitted Fresnel equation to the experimentally measured frequency spectrum for YTZP loaded at 50 N.

4. Results

In order to obtain a direct stress optic coefficient, the refractive index, $n$, was plotted as a function of applied stress for each sample at each load. A linear fit was then applied to each data set in order to determine the stress optic coefficient. YTZP and PTFE are described separately in the following two sections. YTZP samples showed a typical absorption coefficient of 2.5 cm$^{-1}$, while PTFE did not show a measureable value.

4.1 YTZP ceramic

The refractive index measured at each load is plotted against the applied stress for YTZP samples in Fig. 4(a). Stress was calculated by dividing the load by the initial, unloaded cross-sectional area of each specimen, with the initial thickness measured by the zero load fit (described in the previous section), and the width measured with the micrometer. Three YTZP samples were tested and each sample was measured twice. Figure 4(a) shows excellent repeatability for each sample, but a small offset exists between the respective samples.

![Figure 4(a)](image)

(a)

![Figure 4(b)](image)

(b)

![Figure 4(c)](image)

(c)

Fig. 4. Refractive index plotted against applied stress for YTZP. (a) Two repeated measurements for three samples (numbered). (b) Removal of offset in refractive index between samples due to thickness measurement uncertainty. Both (a) and (b) assume constant specimen thickness in the analysis. (c) Comparison between analyses assuming constant or variable specimen thickness.
We believe these small offsets are explained by the uncertainty in the measured thickness of each specimen, rather than by any significant difference in material properties caused by the manufacturing process (including grinding). To demonstrate this, Fig. 5 shows the results of a simulation in which small changes in the assumed sample thickness modify the refractive index values calculated by the non-linear least squares fit to the Fresnel equations, i.e. a numerical estimate of $dn/dl$. For the simulation, transmission curves were calculated using Eq. (4) taking the average refractive index value for the zero stress points in Fig. 4(a) (5.128) and thickness values between 0.653 and 0.664 mm with a spacing of 0.1 µm. A non-linear least squares fit to the Fresnel equation was then applied to each of the calculated transmission curves to recalculate the refractive index, but using a fixed thickness of 0.653 mm. Figure 5(a) shows the difference between the refractive index recalculated from the fit and the nominal value used in calculating the transmission curves, plotted against the difference between the thickness used to calculate each transmission curve and the fixed thickness used in the fit, from which $dn/dl = -0.0078 /\mu m$. The refractive index offset between the three samples from Fig. 4(a) of approximately 0.03 therefore corresponds to a potential difference in thickness of just 3.8 µm, well within the uncertainty of the thickness measurement.

![Simulated refractive index change vs. thickness](image)

**Fig. 5.** Results of a simulation of the change in refractive index as a function of thickness, $dn/dl$ for YTZP and PTFE.

Clearly, small thickness-related offsets in the absolute refractive index are not important when calculating the stress optic coefficient for which the change in refractive index with a change in stress is important. Therefore, the refractive index offset between the samples was removed by fitting a cubic function,

$$f(\sigma) = a\sigma^3 + b\sigma^2 + c\sigma + d \quad (6)$$

to each data set in Fig. 4(a). A cubic function was chosen because viscoelastic behavior results in non-linear response of the refractive index to applied stress [21]. While YTZP (Fig. 4) remained elastic and therefore has a linear refractive index response to applied stress, PTFE will be shown in the following section to exhibit a viscoelastic behavior (Fig. 6). Therefore, a cubic function was chosen to use the same processing for both cases. A new function $f_0(\sigma)$ was fitted to the mean refractive index at each stress. The scalar term $d$ for each data set was then varied to minimize the least-squares difference between the fitted curves and $f_0(\sigma)$ while the constants $a$, $b$ and $c$ were kept fixed. Figure 4(b) shows the normalized refractive index where the variation in the term $d$ for each data set has been subtracted from each point in the data set. This process maintains the functional shape of each sample curve, but removes the refractive index offset due to measurement uncertainty in the specimen thickness.
Finally, Fig. 4(c) shows the mean refractive index at each stress measurement point from Fig. 4(b) plotted against stress. Assuming a constant sample thickness under loading, i.e. engineering stress, the direct stress optic coefficient calculated from a least-squares linear fit is $0.1285 \pm 0.0155 /\text{GPa}$. This line is shown in Fig. 4(c) and also by a dashed line in Fig. 4(a) and 4(b).

In practice, the sample cross-section decreases slightly as the load increases, i.e. true stress. At each load, the instantaneous sample thickness and width can be estimated from,

$$\Delta d = \frac{d \nu P}{AE} \tag{7}$$

where $d$ is the initial dimension (thickness or width) at 0N load, $\nu$ is Poisson’s ratio, $E$ is the tensile modulus, $P$ is the applied load and $A$ is the initial cross-sectional area. Equation (7) is valid when a material is in the elastic regime. Tensile tests showed that YTZP was elastic over at least the applied load range (0 to ~27 MPa). In a repeat calculation, the instantaneous sample thickness at each load was also used in the non-linear least squares fit to the Fresnel equation used to calculate the refractive index: the fit was repeated at every load for each specimen and the thickness was also allowed vary, in addition to the refractive index and absorption coefficient. Figure 4(c) shows the mean refractive index calculated at each load allowing for changes in the specimen thickness, which introduces changes both to the value of $n$ via the fit to the Fresnel equation and to the stress via the instantaneous cross-sectional area. The direct stress optic coefficient calculated from a least-squares linear fit is $0.1362 \pm 0.0155 /\text{GPa}$. The difference between the variable and constant thickness analyses is small, which is to be expected due to the high tensile modulus of YTZP.

### 4.2 PTFE

Figure 6(a) shows the measured refractive index plotted against applied stress for the PTFE samples that were cut with the long specimen axis perpendicular to the extrusion direction of the bulk sheet. Six independent samples were tested. Unlike YTZP, plastic deformation meant that PTFE samples could not be used for repeated tests. Once again, the results show a vertical offset in the measured refractive index between samples, which can be explained by a combination of small errors in the measured sample thickness and variations in the manufactured sample thickness. To demonstrate this we used the simulation of $dn/dl$ as described in the previous section. Figure 5 shows the change in refractive index versus change in thickness from the simulation with PTFE. The average refractive index for the zero stress points in Fig. 6(a) (1.4013) and thicknesses between 2.083 and 2.169 mm with a spacing of 10 µm were used to calculate the transmission curves, and the refractive index was recovered with the non-linear least squares fit using a fixed thickness of 2.083mm. From Fig. 5, $dn/dl = -0.63 /\text{mm}$. The difference in measured refractive index between the six samples from Fig. 6(a) of approximately 0.046 therefore corresponds to a potential difference in thickness of 73.5 µm, which is somewhat larger that the thickness variation seen for YTZP. The PTFE specimens were manufactured from standard extruded sheet, while the YTZP specimens were ground to their final thickness. We typically measured a 21 µm thickness variation across the PTFE specimens, which produced a larger uncertainty in the thickness recovered by the fitting procedure. Despite this offset in absolute refractive index values, Fig. 6(a) shows that the shape of each trace is highly repeatable for each specimen, and once again it is the change in refractive index with a change in stress that is important when calculating the stress optic coefficient. PTFE results show strong viscoelastic behavior above ~ 4.5 MPa, approximately 200 N for the sample dimensions used here. Hence the offset between the measured refractive index due to uncertainty in measured sample thickness was removed by a similar fitting process as described in the previous section, but applied to the data points predominantly linear elastic region below 4 MPa only.
Figure 6(b) shows the normalized refractive index where the constant term in the fit for each data set has been adjusted to minimize the least-squares difference between the fitted curve and \( f_\sigma \), while the constants a, b and c were kept fixed. As before, this process maintains the functional shape of each sample curve, but removes the refractive index offset due to measurement uncertainty in the specimen thickness. Finally, Fig. 6(c) shows the mean refractive index at each stress measurement point from Fig. 6(b) plotted against stress. Assuming a constant sample thickness under loading the direct stress optics coefficient was calculated to be \(-1.3873 \pm 0.1047\) /GPa assuming constant thickness of the specimens in the analysis. This linear fit is shown in Fig. 6(c) and also by a dashed line in Fig. 6(a) and 6(b). Figure 6(c) also shows the direct stress optic coefficient calculated assuming variable thickness in both the fit to the Fresnel equation and instantaneous stress, as described in the previous section. The direct stress optic coefficient for PTFE was only calculated in the elastic regime, and therefore the use of Eq. (7) for the variable thickness analysis is valid. In this case the direct stress optic coefficient was found to be \(-0.1979 \pm 0.1125\) /GPa. Due to the comparatively low tensile modulus of PTFE of 500 MPa, changes in thickness under load are more significant compared to YTZP.

Finally, Fig. 6(d) shows results for six specimens cut with their long axis parallel to the extrusion direction of the bulk PTFE sheet. Direct stress optic coefficients of \(-1.1117 \pm 0.1239\) /GPa and \(0.0785 \pm 0.116\) /GPa were obtained for the constant thickness and variable thickness analyses, respectively.

5. Discussion

YTZP is the main focus of this paper, due to its use in high value manufacturing applications such as thermal barrier coatings and medical implants. The direct stress optic coefficient was
found to be $0.1362 \pm 0.0155 \text{ /GPa}$ in the range 260-380 GHz and was linear for stresses up to 27 MPa. These measurements are the first demonstration of stress induced birefringence in a ceramic, and identify the feasibility of quantitative sub-surface stress measurements in structural ceramics and ceramic coating systems in the future. In addition to two-dimensional stress distributions, depth resolved measurements might be obtained by methods such as confocal imaging or tomography [21–23].

Our interest in PTFE arose because it was the only material for which a stress optic coefficient at GHz or THz frequencies has been reported, but there is clear disagreement in the literature on its value as noted in the Introduction. The direct stress optic coefficient was found to be linear for stresses up to ~4.5 MPa, although the actual value depended on the direction of cutting the specimens from bulk material. Allowing for changes in the thickness of the PTFE specimens, i.e. true stress, our results indicate that the stress optic coefficient is only significant once non-linear, viscoelastic deformation has taken place. It has been suggested that the change in refractive index follows a cubic form for viscoelastic materials [21] and that a strain optic law should be used [12,13], which could be explored for PTFE in future research. Ignoring the change in specimen thickness produces a relatively large change in the measured stress optic coefficient due to PTFE’s relatively low Young’s modulus. Future work to corroborate the stress optic coefficient PTFE reported herein should incorporate effects due to change in specimen thickness and specify specimen parameters such as extrusion direction. Clearly the manufacturing process of the material also affects the measured value.

6. Conclusion

In this paper, the direct stress optic coefficient for YTZP was measured to be $0.1362 \pm 0.0155 \text{ /GPa}$ in the range 260-380 GHz. YTZP samples showed a linear relationship between refractive index and stress, across the entire loading domain up to 27 MPa. For PTFE, the direct stress optic coefficient was found to be linear for stresses up to ~4.5 MPa, although the actual value depended on the direction of cutting the specimens from the bulk material. Above ~4.5 MPa, PTFE did not obey the linear stress optic law. Measurement of the stress optic coefficient for YTZP has paved the way for work in defect detection in high value manufacturing components such as thermal barrier coatings and medical implants. Results in this paper provide information necessary to conduct quantitative photoelasticity measurements of visibly opaque materials, at sub-millimeter wavelengths. Combined with confocal imaging, 3D stress images of manufactured components could be obtained.

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