Technique to measure change in birefringence under shock compression

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Abstract. A technique has been developed to measure the change in birefringence along the axis of shock propagation, probing the relative refractive indices of the material perpendicular to shock propagation. Experiments were performed on calcite single crystals and the results compared to previous literature studies on calcite quasi-static behaviour. Interface velocities are determined using fibre based homodyne Photon Doppler Velocimetry (PDV) operating at 1550 nm whilst the birefringence technique uses free space 532 nm optics. A change in birefringence of $\Delta n = 0.0029 \pm 0.0001$ was observed. This was higher than the predicted change found using a hydrostatic model based on previous studies.

1. Introduction
Anisotropy, defined as a difference in material properties with direction, is present to a degree in all solid-state materials. One anisotropic material property is birefringence, which occurs due to the difference in electronic structure creating a material with a refractive index which is different in one orientation to another. A birefringent material has two identical principle refractive indices (the ordinary refractive indices $n_o$) and one different (the extraordinary refractive index $n_e$). The difference between these is termed the birefringence $n_e - n_o = \Delta n$. In crystalline materials the electronic structure has the same symmetry as the crystal structure, so a birefringent material must have triclinic or hexagonal symmetry. Measurement of birefringence can be a semi-quantitative indicator of strain in a material and is used to test plane-stress models for statically compressed materials. In the shock regime birefringence has been used as a means to visualize shock waves and recent measurements [1] have determined birefringence of sapphire using a Velocity Interferometry System for Any Reflector (VISAR).

Due to the difficulty in modelling anisotropic materials and observations that some anisotropic materials behave isotropically under shock conditions, it is desirable to investigate the validity of using isotropic models to describe materials that are clearly highly anisotropic in the static regime. Birefringence measurements provide a clear indication of whether a material is behaving isotropically or not and hence was the motivation for this project. This study demonstrates a technique for quantifying the change in birefringence under shock compression in calcite single crystals. Calcite is used as a model material because the elastic properties are well characterised under shock compression and the material has a strong initial birefringence. Section 2 outlines the quasi-static elastic and optical model used for calcite to interpret the results in section 4.
Figure 1. Longitudinal stress $\sigma_{xx}$ particle velocity $u_p$ Hugoniot for calcite [2] and copper assuming a 340 m s$^{-1}$ impact. The flyer-driver impact is marked as state A. The driver Hugoniot (copper) is taken to be the release isentrope, a close approximation at low stresses. The point where the driver and calcite Hugoniots cross, marked B, indicates the state of the material behind the shock propagating from the copper calcite interface surface. Due to state B being above the calcite HEL, but below the velocity required to overdrive the shock, there was an elastic precursor followed by a plastic shock.

Figure 2. The calcite plate impact experiment at impact velocity $v$. An initial birefringence of $\Delta n_0$ is used for the unshocked material. An elastic precursor is travelling at $C_L$, the shock wave at $U_s$ and the calcite-copper interface at $u_p$. Each wave introduces a step function in the birefringence. The elastic precursor has a birefringence of $\Delta n_1$ and the shocked state has a birefringence of $\Delta n_2$.

2. Background and Model
A simple isotropic model was used to represent the calcite $\langle 10 \overline{1} 0 \rangle$ behaviour under shock compression. A Tresca strength model was used with longitudinal sound speed $C_L = 7.34$ km s$^{-1}$, bulk modulus $K = 129.35$ GPa, density $\rho = 2.71$ g cc$^{-1}$, and a Hugoniot Elastic Limit (HEL) pressure of 1.15 GPa [2, 3]. The authors of reference [2] found that for this orientation a particle velocity of 1.4 km s$^{-1}$ was required to overdrive the shock. Given the expected impact speed is 340 m s$^{-1}$ the expected longitudinal stress $\sigma_{xx}$, found using figure 1, is above calcite’s HEL. This created a two-wave structure in the sample, an elastic precursor followed by a plastic shock.

The optical model used for calcite assumes a step pressure wave propagating through the material leading to a discontinuity in the birefringence. The pressure $P$ is found from the average of $\sigma_{xx}$ and transverse stress. The refractive indices are dependant on pressure, as measured by [4], so a static high-pressure model was implemented. The predicted pressure state behind the wave is used to predict the birefringence behind the wave. The values of $n_o$ and $n_e$ are given by the linear pressure relation $n_o = n_{o0} + C_o P$ and $n_e = n_{e0} + C_e P$ where $C_o = 0.050$ GPa$^{-1}$ and $C_e = 0.044$ GPa$^{-1}$ and the atmospheric refractive indices of calcite are $n_{o0} = 1.658$ and $n_{e0} = 1.486$.

In a birefringent material light polarised along the $n_e$ and $n_o$ directions will have different
wavelengths, hence the emerging ray will have a different phase depending on whether it was polarised along the \( n_e \) or \( n_o \) directions. Light polarised at an arbitrary angle \( \theta \) to \( n_e \) will have a component in \( n_e \) and \( n_o \) given by standard vector calculations. The phase difference \( \phi \) between the \( n_e \) and \( n_o \) polarised components after a ray of wavelength \( \lambda \) has propagated a distance \( l \) through the sample is given by equation (1). When the ray emerges, summing the \( n_e \) and \( n_o \) components will give the polarisation state of the emerging ray.

\[
\phi = \frac{2\pi}{\lambda} l \Delta n
\]  

(1)

As the experiment is expected to generate both an elastic precursor and a shock wave, the optical model must be extended to include multiple waves. The total phase difference \( \Phi \) was found by treating the material between each wave as a single birefringent sample creating a phase difference of \( \phi \), and then summing over all wavefronts as in equation (2). Substituting in equation (1) gives equation (3). In order to measure \( \Phi \) circularly polarised light was used so that the intensity of the \( n_e \) and \( n_o \) components are identical and independent of the angle \( \theta \).

\[
\Phi = \sum_{i=1}^{k} \phi_i
\]  

(2)

\[
\Phi(t) = \frac{2\pi}{\lambda} \sum_{i=1}^{k} l_i(t) \Delta n_i
\]  

(3)

3. Experimental Method

3.1. Birefringence

The birefringence of the sample was probed perpendicular to shock propagation. A circularly polarised beam of 532 nm was used as the probe and the final polarisation state investigated with a Thorlabs polarising beam splitting cube, orientated with the S and P polarisations at 45 degrees to the calcite \( n_o \) and \( n_e \) directions. The ratio of detected S to P polarisation is dependant on the polarisation state of the light incident on the cube, which in turn is dependant on the phase difference between the \( n_o \) and \( n_e \) polarised components. Thorlabs DET10A fast (1 ns rise time) unamplified photo detectors were used with a 10 V reverse bias and terminated with a 50 Ohm resistor in the oscilloscope. A Photon Doppler Velocimetry (PDV) probe was made collinear with the birefringence probe using a suitable dichroic mirror. A photograph and schematic layout of the diagnostic is given in figure 3.

3.2. Target Design, PDV, and Plate Impact

The target was an Iceland Spa calcite single crystal sample 13 mm in diameter and 2 mm thick cut with the basal plane perpendicular to the faces (i.e. \( \langle 1\overline{0}10 \rangle \) direction perpendicular). The sample was cut and polished by Crystran Optics and found to have a thickness of 2.067 ± 0.001 mm and the flat faces were parallel to one micron. A 500 nm aluminium layer was thermally evaporated onto one surface of the sample in order to get a reflective boundary with negligible transmission. The target was affixed to a 16 mm diameter, 1.903 ± 0.001 mm thick copper driver plate lapped flat and parallel to 1 micron. The buffer and the calcite were bonded together with low viscosity epoxy, and the glue layer thickness was measured to be less than 5 microns.

Imperial College London’s mesoscale light gas gun facility was used, and the target aligned perpendicular to the barrel to 2 mrad. A light gate measured the impact velocity to be
Figure 3. (a) Photograph and (b) Schematic of the birefringence diagnostic. The green line marks the path of the 532 nm Verdi and the red marks the 1550 nm beam emitted from the fibre probe. The purple line is the combined beam. The lambda / 4 plate is a 532 nm quarter wave plate used to change the linear polarised light to circularly polarised.

Figure 4. The signal data (a) compared with modelled data (b), the line marked Fit is a sine wave with the same frequency in (a) and (b), though the amplitude has been scaled in (a). The oscillations in polarisation are faster in the data of (a) than in the prediction of (b). As shock velocities were taken from figure 5, the change in birefringence must be larger than expected.

$340 \pm 20 \text{ m s}^{-1}$. The diagnostics were all fed into one 2.5 GHz bandwidth scope sampling at 10 Gigasamples per second for 500 $\mu$s. One channel was used for a homodyne fibre based PDV system operating at 1550 nm wavelength [5], and the remaining three used for the S, P and Beam Intensity Monitor (BIM) detectors.

4. Results
The normalised signals from the birefringence detectors are shown in figure 4 (a). The shock Time-Of-Arrival (TOA), determined from the PDV data in figure 5, is marked. However, due to the length of fibre used for the PDV detector the TOA on the PDV oscilloscope trace does not match up with the TOA on the birefringence oscilloscope trace. There is approximately an extra 8 metres of fibre introducing an approximate 40 ns time delay on the PDV signal compared to the birefringence signal. This optical path length difference was not fully calibrated in the
Figure 5. Spectrogram of PDV data. The black trace indicates the mean value of a fitted Gaussian to the peaks in the PDV power spectrum. Region B is caused by reflection from the copper calcite interface surface. Region C is the precursor wave reaching the free surface whilst region E is plastic wave breakout. The feature at D is signal drop out. The spectrogram was constructed with a Fast Fourier Transform (FFT) size of 1024 points using a Hann window of size 256 points and an overlap of 160 points.

original experiment, so the exact arrival time of the shock wave on the birefringence data is not known. As this systematic error was not properly characterised, the time axis in figure 4 (a) has not been corrected. A line marking the estimated time difference of 43 ns has been added. This estimate was reached by assuming that the slow change in polarisation state initially observed (marked A in figure 4 (a) ) corresponds to region A in figure 5 and the faster oscillations marked region B correspond to the shock arrival in figure 5.

The oscillations in region B of figure 4 (a) have been fitted to a sine wave and compared to a predicted signal in figure 4 (b) generated from the model outlined in section 2. The $u_p$, $U_s$ and $C_L$ values used for this model were experimentally determined from the PDV data in figure 5, and the pressure found by averaging the transverse and longitudinal stress calculated from the calcite (1 0 1 0) Hugoniot. This pressure was used to predict $\Delta n_1$ and $\Delta n_2$, and the predicted signal found by calculating $\Phi$ from equation (3) and resolving into S and P components with an ideal $\theta$ of 45 degrees. There is a noticeable static component to the polarisation, most likely due to a reflection from the surface of optical elements and particularly the calcite window which was not coated with an anti-reflective layer. The fitted sine wave frequency yielded a change in birefringence of $0.0029 \pm 0.0001$ though this technique cannot determine whether this is a positive or negative change at present.

Values for $u_p$ were determined using the PDV data shown in figure 5, $U_s$ and $C_L$ values were calculated by using the known sample thickness and the TOA data from the raw PDV signal. The initial peak, marked B, from light reflected at the aluminium layer (copper calcite interface) was broad, likely due to the two refractive indices of calcite. A correction to the observed velocity to adjust for the refractive index of the material has not been applied. The free surface trace has an elastic region (marked C) followed by the plastic shock, marked E. Shock transit times gave a shock velocity for the elastic precursor of $7.50 \pm 0.11$ km s$^{-1}$ and a Eulerian $U_s$ of $4.74 \pm 0.05$ km s$^{-1}$ for the shock. Assuming $u_p$ is half the free surface velocity (valid at low pressures), then the corresponding $u_p$ was found to be $114 \pm 4$ m s$^{-1}$ and $224 \pm 4$ m s$^{-1}$ respectively. A $u_p$ of 224 m s$^{-1}$ is lower than the observed velocity in region B, as expected for
a measurement made through a window material.

5. Discussion
In this experiment the impact velocity generated a state above the HEL leading to a two-wave structure in the material. At present it is not possible to separate the two absolute birefringence values from the raw data. Due to the loss of signal from the birefringence measurement after about 40 ns, it is likely that the second wave caused fracturing in the material, rendering it opaque. It has been assumed that birefringence data recorded is representative of the precursor, however more experiments at lower impact velocity are required to verify this.

The PDV data agreed well with previous studies by [2, 6], and the particle velocities were consistent with that expected from figure 1. However the value of \( U_s \) is much lower than the expected bulk sound speed of \( C_b = 6.91 \text{ km s}^{-1} \), calculated from \( K \) and \( \rho \) given section 2, though the experimental result is consistant with experimental results from [2]. This indicates that the Tresca strength model is insufficient, and a more appropriate strength model will be developed.

6. Conclusions
- Calcite \( \{1\ 0\ T0\} \) was used in a plate impact experiment on copper at \( 340 \text{ m s}^{-1} \) and a change in birefringence of \( 0.0029 \pm 0.0001 \) measured.
- 1550 nm fibre based PDV was used to measure the velocity history at the same point on the target as the birefringence measurement was taken. This was achieved using a dichroic mirror.

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