

Optical Ptychography of Calcite Thin Films

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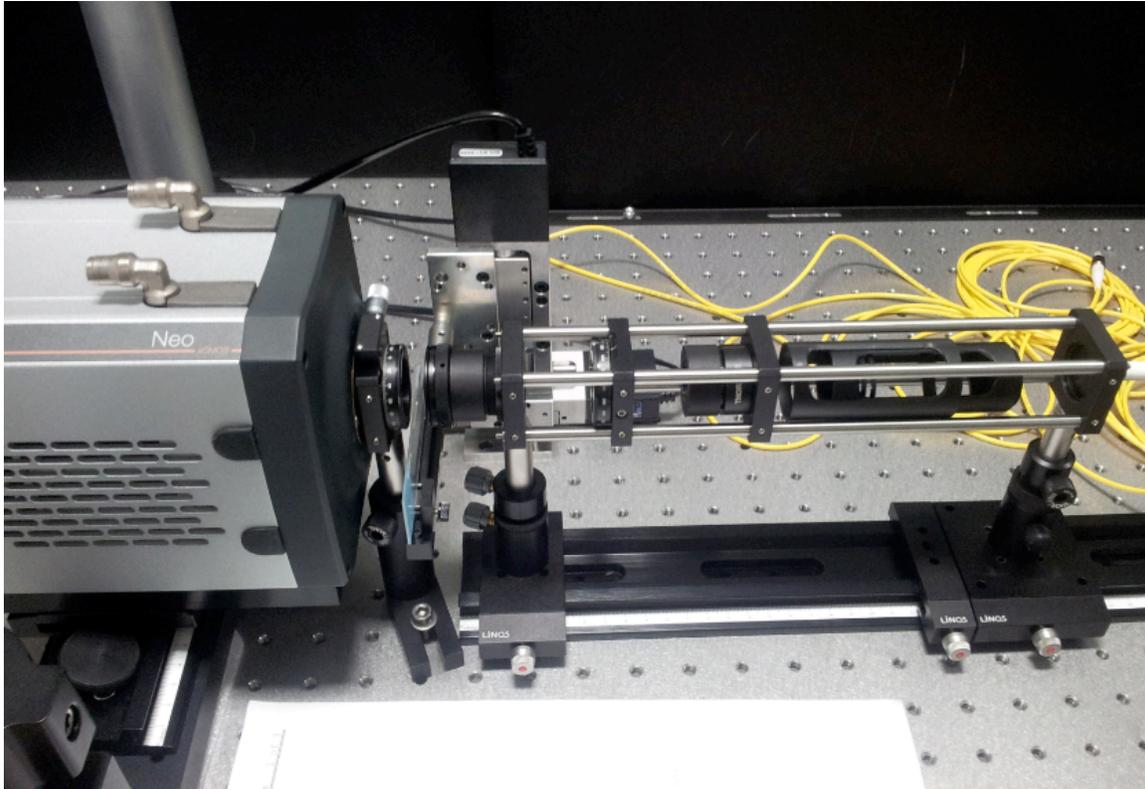


Figure 1: Set up of ptychography apparatus.

Introduction

The aim of this project was to use a recently developed imaging technique known as optical ptychography to examine thin films of calcite crystals grown using a method similar to that used by collaborators at Leeds University. Ptychography is a coherent diffraction imaging technique that works by recording overlapping diffraction patterns and then running an iterative algorithm to create a reconstruction of the object's transmission function. In doing this it is able to overcome the phase problem associated with the measuring of a diffraction pattern, giving good numerical values for the object's phase. A further advantage of the technique is that no lenses are required to create the image, meaning that the resolution of the final image is limited by diffraction effects only. This is particularly useful when considering extending the technique to x-ray imaging where the use of optical components has a more significant effect on the image resolution and are much more difficult (and expensive) to fabricate.

Ptychography

For a thin object, we can define a complex 2D transmission function $O(\mathbf{r})$, which along

with knowledge of the illuminating probe function $P(\mathbf{r})$ has a relatively simple Fourier transform relationship with the measured diffraction pattern. However when an experiment is carried out, it is only possible to measure the intensities of the diffraction pattern, and so all the information regarding the phase is lost. Consequently it is not possible to simply apply an inverse Fourier transform to obtain the transmission function of the object. This problem is overcome by sampling the object with the probe at many different positions, but crucially setting it up so that the positions partially overlap. The algorithm then works by making a guess at the transmission function, which is gradually improved via applying Fourier transforms back and forth and applying the appropriate constraints - namely that the intensities of the diffraction pattern must match the recorded intensities and that the transmission function in all areas of overlap must be consistent with all of the recorded diffraction patterns. The algorithm also works to refine the illumination function so that only a good knowledge of $P(\mathbf{r})$ is required, not a perfect one.

At present, the technique appears to be more robust than the "single shot" CDI techniques where the object is reconstructed from a single diffraction pattern using a support constraint, as these require either a very good knowledge of the probe, or a probe which is larger than the object being imaged to remove any ambiguity whereby multiple transmission functions can all satisfy the given constraints. If this is the case, the algorithm cannot reliably converge to the correct solution. One disadvantage with ptychography is the time it takes to record a sufficient number of diffraction patterns (usually at least 25 are needed to be able to consistently reconstruct a single image, although we have tended to use 81), which limits its applicability to imaging any dynamic systems. It also requires that the set up of the equipment is very stable.

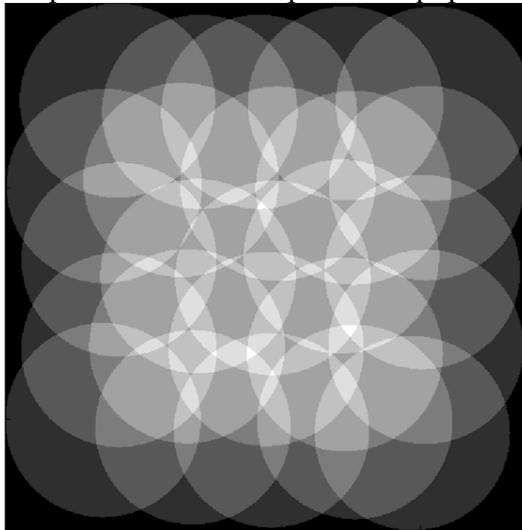


Figure 2: Example of an overlap map of the different positions that a probe illuminates an object, each producing a diffraction pattern which is measured. The object is then reconstructed using an iterative algorithm and the constraint that the object in the area of overlap must be consistent with producing the diffraction pattern from each probe.

Calcite

Calcite is the most stable polymorph of Calcium Carbonate, and is of interest in optics

mainly due to the large birefringence it exhibits. It is also widely studied in the field of Biomineralization as many animals are able to exert great control over the crystal location, size, orientation as well as the actual state of the Calcium Carbonate that is produced. Biomimetic engineers wish to achieve a greater understanding of how these organisms do this, and extrapolate the knowledge for applications in other areas such as materials science.

Recently it has emerged as a popular opinion that many biominerals may be formed via an amorphous precursor pathway, which strongly influences the properties of the crystals that form. In particular, the Leeds collaborators have been investigating how adding certain polymer additives can temporarily stabilize an amorphous phase of CaCO_3 with certain fluidic properties, before eventually crystallizing into the more thermodynamically stable Calcite.

Although there is now good evidence that this is indeed what happens, one of the aims of this project was to watch the growth of the crystals in situ by imaging them as they developed. By using different linear polarizations for the probe along with the accurate phase measurements offered by ptychography it would be possible to see when the birefringent crystals emerge, and perhaps gain some insight into the process at work prior to this. However due to various problems such as not being able to consistently grow the crystals as desired, and not having control over where the crystals form we have not been able to do this yet.

Results

Figure 2 shows images of some of the crystals grown, using both standard and polarized light microscopy. It is quite clear that there are many single crystal domains which have formed with different orientations (and possibly varying thicknesses also), which is why they appear as different colours under the polarized microscope. This is believed to happen because a film of the liquid amorphous CaCO_3 forms on the surface, before nucleation starts randomly at different sites across the film.

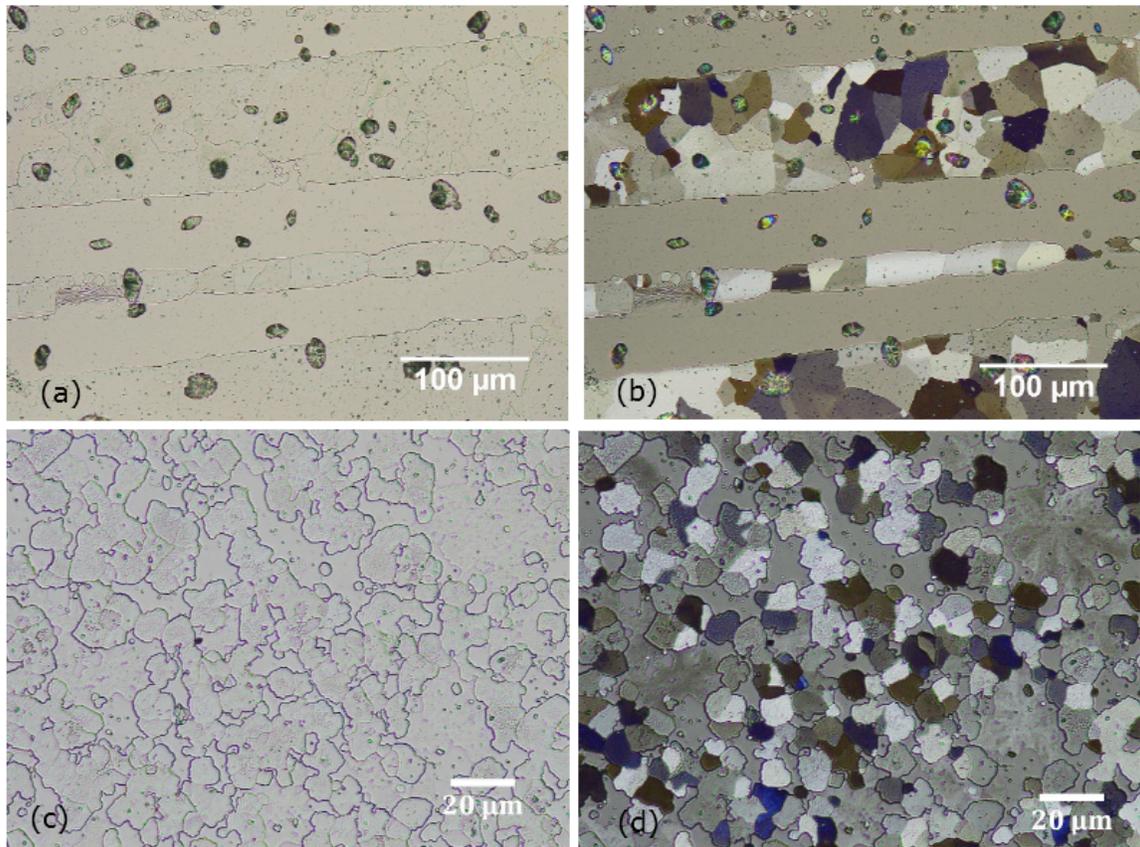


Figure 3: Images of some of the crystals grown in different areas, with a normal microscope ((a),(c)) and a polarized microscope ((b),(d)).

Figures 3 and 4 show reconstructed images of a similar area to fig 2(a) and 2(c) using the ptychography set up. This provides both an intensity image of the object as well as a phase map and provides a numerical value for each pixel. Figure 3 is taken with a vertical polarizer before and after the sample, and figure 4 with almost crossed polarizers before and after (the reason they're almost crossed and not completely perpendicular is that there must be some light that can pass through areas of no sample, or else a phase cannot be meaningfully defined there).

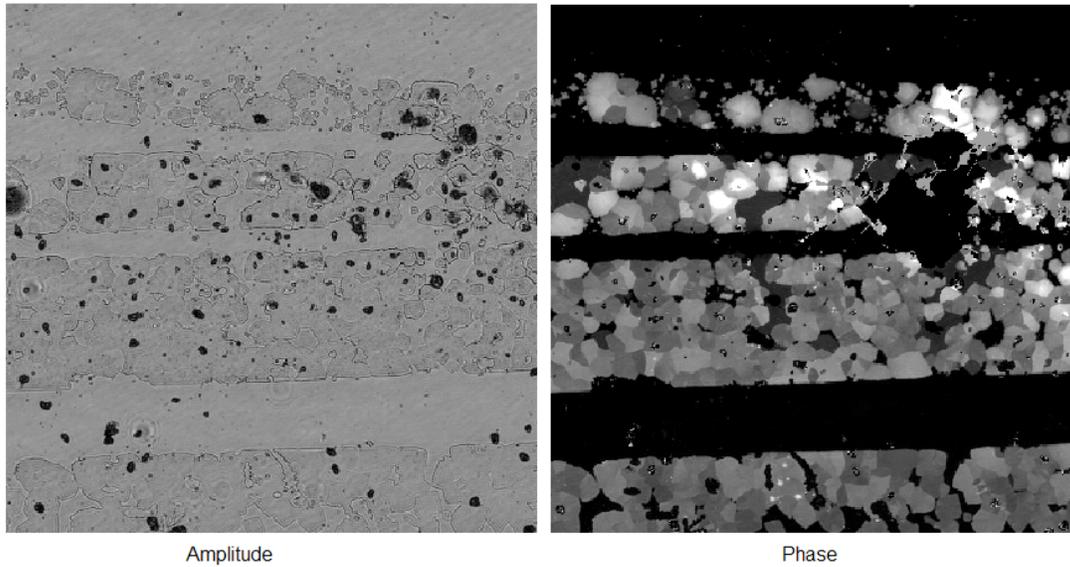


Figure 4: Reconstructed image using the ptychography technique, which provides both an intensity image and a phase image.

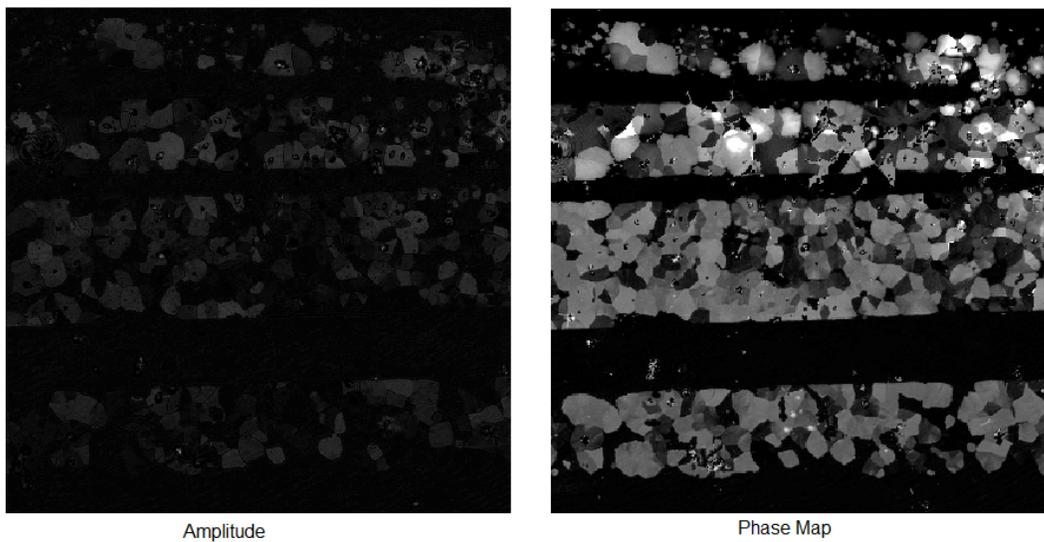


Figure 5: Another reconstructed image of the same area using the ptychography set up, with almost crossed polarisers (one before the sample, one after).

Orientation of Calcite Crystals

Another thing we wanted to do was see if we could find a way to use the ptychography technique to determine the orientation of the calcite crystals, as this could be useful in further investigations into how it may be controlled.

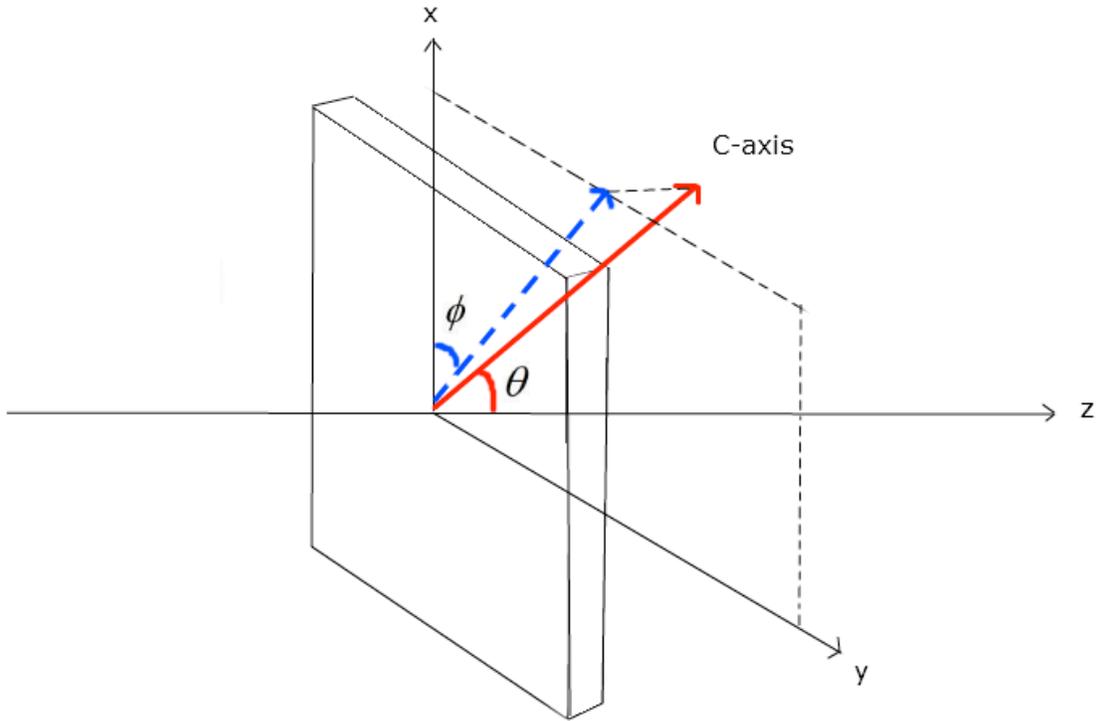


Figure 6: The probe travels in the z-direction, and we wish to calculate the values of θ and ϕ to see how the Calcite c-axis is oriented. θ is the angle between the c-axis and the propagation direction, and ϕ is the angle between the projection of the c-axis onto the x-y plane and the x-axis.

We approached this problem using the methods of Jones calculus. The probe is initially linearly polarized so that we know the initial Jones vector, and assuming normal incidence we can calculate the resultant Jones vector for propagation through the Calcite, with c-axis at θ to the propagation direction, and its projection onto to x-y plane at angle ϕ to the x-axis as follows:

$$\begin{pmatrix} |E_x| e^{i\phi_x} \\ |E_y| e^{i\phi_y} \end{pmatrix} = \begin{pmatrix} \cos(\phi) & -\sin(\phi) \\ \sin(\phi) & \cos(\phi) \end{pmatrix} \begin{pmatrix} e^{\frac{i2\pi d(1-n_e(\theta))}{\lambda}} & 0 \\ 0 & e^{\frac{i2\pi d(1-n_o)}{\lambda}} \end{pmatrix} \begin{pmatrix} \cos(\phi) & \sin(\phi) \\ -\sin(\phi) & \cos(\phi) \end{pmatrix} \begin{pmatrix} E_{x,init} \\ E_{y,init} \end{pmatrix}$$

where:

$$\frac{1}{n_e(\theta)^2} = \frac{\cos(\theta)^2}{n_o^2} + \frac{\sin(\theta)^2}{n_e^2}$$

Using two different initial polarizations, and then measuring both the x and y components of the resulting polarizations (4 measurements in total), it should be possible to recover ϕ , θ and d . We have yet to test this method properly, which ideally would be done using a

sample with known orientation, however from the simulations we've carried out we do think that the technique can work. Despite this, there may be problems with the accuracy of the data we require. In particular the ptychography method may not give us an accurate enough measure of the amplitudes $|E_x|$ and $|E_y|$, and also it is clear that because θ must be calculated from $n_e(\theta)$ which can only vary from 1.486 to 1.658, an accurate θ requires very accurate phase and amplitude measurements.

Conclusion

Overall I have really enjoyed my time at Harwell working with a great team on a very interesting subject. I have learnt a lot about how research is carried out and what to expect if I continue down that path (which I hope to do so), as well as acquiring a lot of useful experimental and computational skills and meeting a lot of very nice and interesting people along the way. The project itself has been both enjoyable and challenging and a great way to spend the summer!