ESRF	**Experiment title:** Phase-imaging of Periodically-Poled Crystals of the $KTiOPO_4$ Family	**Experiment number**: HS-709
Beamline:	**Date of experiment**: original dates from: 24/2/99 to: 27/2/99 then postponed (due to illness) to final dates: 4/6/99-6/6/99	**Date of report**: 25/2/2000
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Report:

Experiments to investigate phase-contrast images, otherwise known as Bragg-Fresnel images[1], of periodically domain-inverted arrays in crystals of $KTiOPO_4$ (KTP) and $KTiOAsO_4$ (KTA) were undertaken during 3 shifts in February 1999 (original scheduled time) and 6 shifts in June 1999 (re-scheduled time). The periodically-poled arrays had periods of 9 microns and 39 microns for KTP and KTA[2], respectively, these choices being determined by the demonstration of real optical devices using these crystals. In each case, the orientation of the induced domain walls was the conventional orientation normal to the [100] crystallographic direction.

1 KTA

The experiments on KTA were conducted using white beam section topography in transmission through the 0.5 mm thickness of the crystal. The Laue method implies that many reflections will be received on the film at any one time: in searching for reflections showing significant phase contrast, this is useful as a large number of reflections can besurveyed at once. Once reflections sensitive to phase contrast had been identified, these were followed as a function of sample to film distance in the range from ~5 cm to 2 m.

The three reflections identified and extensively studied were 1-31 and its harmonic 2-62, 0-31 and its harmonic 0-62 and 1-62. In each case, the phase contrast measured in the experiment was compared with simulations based on the crystal structure of KTA by Mayo, Thomas *et al*. [3]. Now, since KTA is a polar material with space group $Pna2_1$, the position of the origin on the z axis is not crystallographically fixed – it is a "floating" origin. However, in calculating the phase jump between reflections hkl and hk-l across inversion domain walls, as in periodically-poled KTA, the calculated value depends *critically* on the choice of origin. Following the suggestions of Thomas & Glazer[4] about possible origins for inversion twinning in KTP, the origin for phase-jump calculations was placed successively at As(1), As(2), Ti(1), Ti(2) and the calculated position of Ti(2) in the prototypic paraelectric phase. It was found that a self-consistent set of phases which allowed simulation of images in agreement with experiment could only be derived for the choice of origin at As(1). This means that as result of this experiment, we have been able to identify the atomic site through which domain walls normal to [100] are matched, i.e., we have identified the atom that remains effectively static under the twinning operation. We can explain why this occurs using crystallographic principles since this origin choice preserves the continuity across the domain walls of AsO_4/TiO_6 chains along the [100] direction

To our knowledge, this technique, because of its ability to measure absolute phase information, is unique in providing structural detail of this sort about ferroelectric twinning. This work has been completed and written up for publication[5] in Journal of Applied Crystallography.

2 KTP

The experiments on KTP were conducted using both white beam in transmission and monochromatic beam in reflection from the (001) surface. The monochromator used, for the first time for a phase-contrast imaging experiment at ID19, was a metallic multilayer B_4C/Ru with d-spacing 42 Å. Similar analyses to those for KTA described above have been performed on both the white beam and monochromatic beam images with the conclusion that the origin choice in KTP must be the P(2) atom. There are some marginal crystallographic reasons why the preference is for P(2) in KTP rather than P(1) (which would be analogous to As(1) in KTA). However, the case for this needs stronger development and would benefit from further experiments on KTP with different domain wall orientations and periods. One source of concern was the possibility that surface effects, e.g. curvature of the surface, were contributing to an additional phase contrast in the KTP sample. Therefore, the x-ray imaging was supplemented by optical interference microscopy on all the periodically-poled crystals investigated so far using the facility at ESRF in Dec. 99. A significant curvature of the surface of the KTP was not found. Furthermore, these measurements also showed that that periodic surface modulations associated with the inversion domains contribute negligibly to the overall phase contrast – they are swamped completely by the intrinsic phase jumps across the domain walls.

At the time of writing then, the conclusion rests that the domain walls in the particular KTP sample under study are matched through the P(2) atom. Further work is required to confirm this and to explain it convincingly from a crystallographic stand-point. This research is on-going and is the subject of a further application for time on Station ID19.

References

[1] Pernot-Rejmankova, Cloetens, Guigay, Baruchel & Moretti (1998) Phys. Rev. Lett. 81, 3435-3438

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[3] Mayo, Thomas, Teat, Loiacono & Loiacono (1994) Acta Cryst. B50, 655-662

[4] Thomas & Glazer (1991) J. Appl. Cryst. 24, 968-971

[5] “Investigation of a periodically-poled KTA crystal using coherent X-rays” P. Pernot-Rejmankova, P.A.Thomas, P. Cloetens, F. Lorut, J. Baruchel, Z.W.Hu & G. Rosenman. Submitted to J. Appl. Cryst.