COMBINING DIFFRACTION TOPOGRAPHY AND PHASE IMAGING

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This work results from collaboration between ESRF staff members and scientists from other institutes: J. Gastaldi and E. Reinier (CRMC2, Marseille), J.P. Guigay and M. Schlenker (Lab. L. Néel, CNRS, Grenoble), Z. Hu and P. Thomas (Univ. of Warwick), P. Moretti (Univ. C. Bernard, Lyon).

The deliberate combination of phase and Bragg-diffraction imaging has recently led to important results, giving a new impulse to the use of these techniques. The experiments are being carried out on several beamlines: BM5, ID11, ID19 and ID22.

In the classical version of x-ray diffraction imaging ('topography') the single-crystal specimen is set to give Bragg reflection(s), and a detector placed as near as possible records the diffracted beam(s). Contrast shows up whenever the crystal includes features that locally affect the Bragg reflectivity. Thus crystal defects (dislocations, inclusions, ferroelastic domains,...) are made visible and can be characterized. The investigation in transmission of thick crystals often requires restricting one dimension of the beam («section» topography): the image then originates, in a first approximation, from a virtual slice of the crystal. When working at the ESRF an additional parameter can be varied to obtain further information: the 'propagation' distance between sample and detector.

The 'propagation' parameter also allows phase radiography and tomography to be performed in a very simple way. The 'propagation' technique takes advantage of the small size of the x-ray beam sources at ESRF, yielding a very small divergence α of the beam as seen from any point in the specimen. This results in a high degree of spatial coherence, characterised by the transverse coherence length $l_c = \lambda/2\alpha$, where λ is the x-ray wavelength. Local variations in the x-ray optical path-length in the sample, i.e. phase variations across the beam, then produce contrast through Fresnel diffraction. In the case of negligible absorption the image is uniform when the detector (x-ray film in our case) is close to the sample. As the detector is moved further away, contrast builds up, and features on different length scales are brought out more conspicuously depending on the specimen-distance D. The edges of an inhomogeneity of size s are outlined when $D < s^2/\lambda$, whereas interference fringes occur, the image being a hologram, for further increase of D.

The investigation of defects in Al-Pd-Mn single quasi-crystals is a first case where the combination of topography and phase radiography allowed a breakthrough. It is now familiar that quasicrystals, non-periodic solids with long-range order, exist, and produce fine diffraction peaks for x-rays just as do crystals. Therefore Bragg-diffraction imaging is applicable to single quasicrystals too, and it has already produced some information about the structural defects in these materials, with topology quite different from that of crystals. Figures 1a and b show a region of a single quasicrystal slab, imaged in simple transmission, with no strong Bragg reflection excited. In the enlargement (Figure 1a) a clear delineation of internal holes with a shape featuring, just as etch pits in crystals, the icosahedral point symmetry of the material, and elongated precipitates, are observed. Figure 1c shows a topograph of the same region as in Figure 1b. Loop shaped defect images, corresponding to distorted areas, are visible. The geometrical relations between the two, apparently quite different, images of the same object can be determined by recording a phase radiograph with the sample set for a strong Bragg reflection (Figure 1d). Joint use of these images allows to determine the relationships between the holes or precipitates and the complicated defects around them [1].

In conventional Bragg-diffraction imaging, the surfaces of the samples (and/or monochromator) should be strain free, but no specific demand is made on

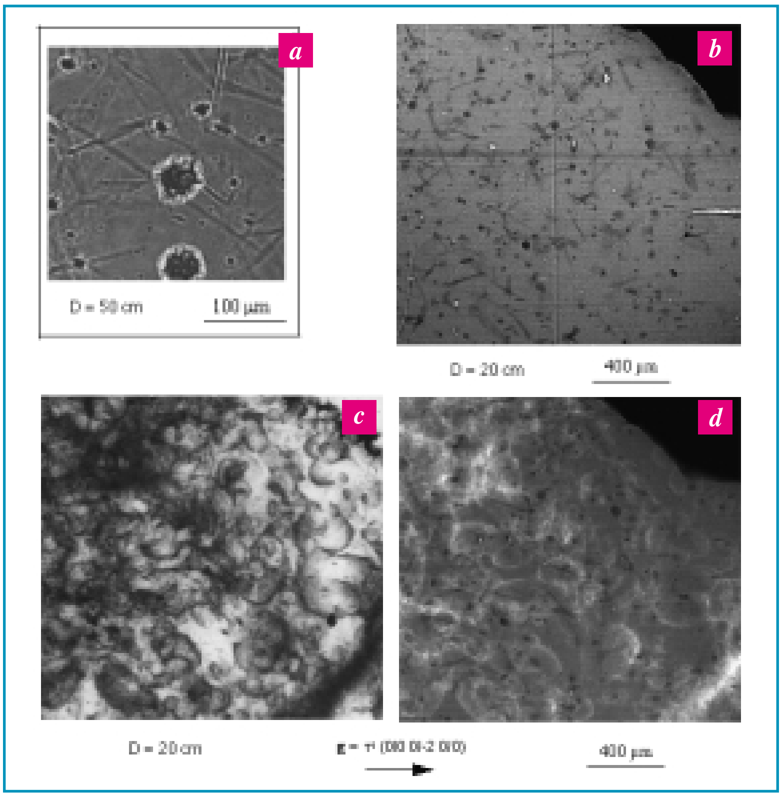

Fig. 1: Images of an Al-Pd-Mn quasicrystal recorded at $\lambda = 0.35$ Å a) phase radiograph (D = 0.5 m). In b), c) an d) the same region of the sample is presented (D = 0.2 m) b) phase radiograph with no strong Bragg diffraction excite; c) topograph (diffracted beam) and d) combined phase radiograph and topograph (transmitted beam); sample set for the τ^3 (0/0 0/-2 0/0) reflection.

their flatness. This is no more true when dealing with high quality crystals and using a coherent x-ray beam. The diffraction image then carries phase information, yielding extra contrast when the optical path length is irregular. Faint scratches on the surface of silicon crystals used as monochromators were shown to produce strong phase images which are not visible in 'classical' topography. The same approach was used to reveal thin oxide strips on reflections where the associated distortion does not produce contrast [2].

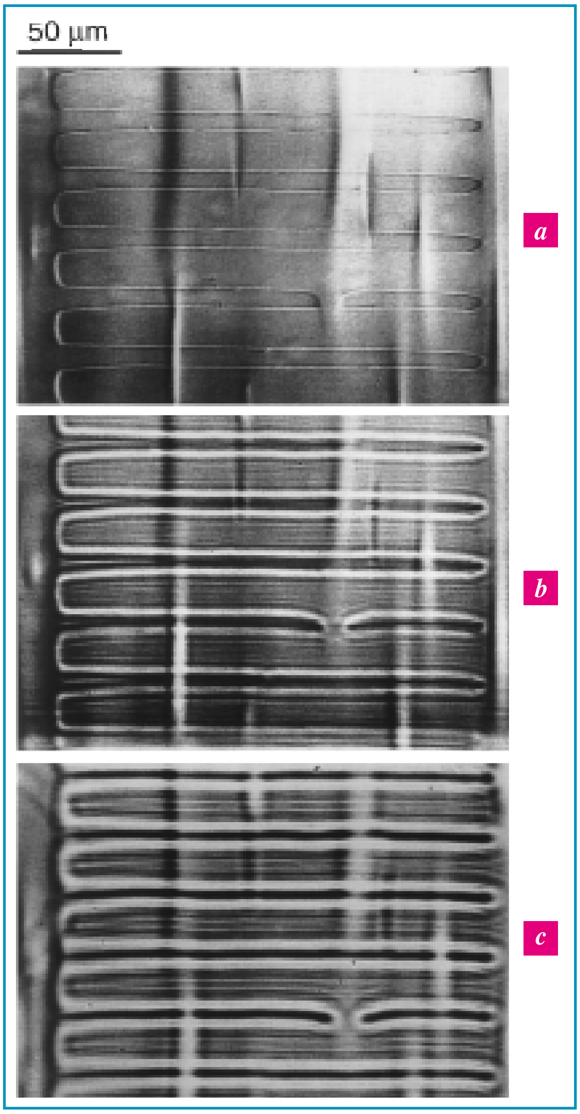

Fig. 2: Images of inversion domain walls in a $LiNbO_3$ crystal recorded using the 006 reflection (reflection geometry), $\lambda = 1$ Å, at different sample-to-film distances D: a) 0.5 cm b) 20 cm c) 50 cm. Note the interference fringes which build up as the distance is increased.

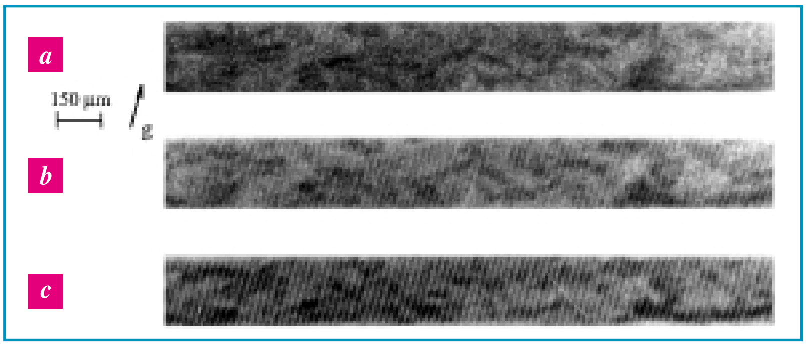

Fig. 3: Section topographs (transmission geometry) of a periodically poled $LiNbO_3$ crystal using the 274 reflection, $\lambda = 0.24$ Å, recorded at different sample-to-film distances D: a) 0.11 m, b) 0.51 m, c) 1.67 m, g is the projection of the diffraction vector on the detector.

The combination of phase and diffraction imaging led to spectacular results with beams Bragg-diffracted by periodically poled lithium niobate crystals. These crystals exhibit large optical nonlinearities and are widely used for electro-optic and acousto-optic applications as well as for optical frequency conversion. Poling produces a periodic spatial distribution of the sign of the nonlinear optical coefficient, crucial for the quasi-phase matching required for optical harmonic generation. When parts of the coherent x-ray wave front diffracted from domain inverted regions are split and subsequently overlap, they interfere [3]. Figure 2 shows a series of images at different sample-to-film distances, in Bragg (reflection) geometry. The fringes show the phase origin of the phenomenon. The inverted ferroelectric domains exhibit structure factors with very similar modules, but different phases. This gives rise to contrast mechanism, used to image the domain distribution in the bulk in Laue (transmission) geometry as shown on Figure 3. Many reflections involve nearly no effect of domain-related lattice distortion. Contrast then mostly originates from the phase shift between the structure factors in the domains. It is thus possible to extract, from images recorded at different distances, a direct measurement of this phase difference [4]. It was on the other hand shown that the contrast of the domain images can be enhanced by the application of an electric field [5].

The above examples show some of the unique possibilities resulting from the combined use of Bragg and Fresnel diffraction imaging techniques. This is clearly an emerging new field. ■

References

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