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Report: The aim of this experiment was to study the structure of the two interfacial regions existing at the α-incommensurate (inc) first order phase transition of quartz at 846 K. These heterogeneous regions produce intense light scattering, known as quartz opalescence, the origin of which remains a puzzling question discussed recently [1].

We use high energy white beam x-ray diffraction in transmission on ID19. The diffracted x-ray beams were recorded mostly with photographic films to observe the whole Laue pattern; some measurements were also performed with the FRELON camera to follow the phase transition in real time. We used our high accuracy furnace, with a temperature control of a few mK, developed for previous optical and x-ray studies [2]. Although the average temperature is very stable, there is a vertical temperature gradient of about 0.1 K/cm which helps to stabilise the interface in the middle of the sample.

Three quartz single crystal samples were studied successively:

Ny, a natural sample plate of 7.5 mm thickness, perpendicular to the Y axis of quartz;

Sx, a, synthetic sample of medium quality from SICN (France); X plate of 4.35 mm thickness;

Sy, a synthetic sample of high quality from SICN (France); Y plate of 4.5 mm thickness.

The Z faces of the three samples were polished to perform a video observation of the phase interface with a white light beam; a He-Ne laser, at an incidence angle of 3° from the Z axis was used to obtain light scattering from the two heterogeneous interfacial region.

In the three samples, some preliminary measurements were performed in the pure inc phase to check the temperature behaviour of the inc satellites. We also get pictures of the rotation domains of the usual inc 3q phase: as observed previously, the domain walls are parallel to the Z axis but with various transverse cross-sections. Sx has the usual lamella structure with thickness of about 0.1 mm ; Ny has rectangular shapes of several mm and Sy a coarse cylindrical structure in the mm range.

Our measurements were performed mostly in the coexistence state with the α and inc phases present together in the sample. The interface can be moved by small temperature variations. The first sample (Ny) was studied in details with photographic films: observations with a thin vertical slit (10x0.1 mm^2) give a general overview of the satellite pattern converging towards lattice Laue spots. However due to the very different intensity of the various features (lattice and satellite spots, diffuse scattering), clear detailed pictures can be obtained only with a thin beam (often from a small horizontal slit of 0.3x0.1 mm^2) and various optimal exposure times (10 to 300 s). A vertical scan at fixed temperature (in the state obtained after heating) showed that the final variation of the inc satellites is discontinuous: starting around 0.03 a* at the lower (and hotter) position, it decreases down to 0.007 a* within 0.1 mm of the inc phase boundary (a* is unit cell vector of quartz in the basal plane of reciprocal space). Simultaneous optical observations along the Z axis show that the inc satellites disappear at the contrasted boundary visible at the colder limit of the SAS region. In comparison with our previous hard x-ray measurements, the use of synchrotron radiation improves the spatial and angular resolutions by one order of magnitude, disclosing unambiguously the final discontinuity of satellite variations. The presence of the discontinuity (with the same value) was confirmed by a slow heating. However upon cooling the satellites move closer to the Laue spot, they are clearly observed at 0.002 a*, where they probably disappear. This ultimate discontinuous behaviour is however ambiguous, as we then observed a diffuse streak with a weak maximum around 0.001 a*. To clarify this point further studies with improved resolution are needed. We observed that the q variations of the satellite is often produced by a discontinuous succession of small patches, probably each with a nearly constant value of q. Similar but less extensive results were obtained in the two other samples.

On the α phase side, diffuse x-ray scattering was observed within 1 mm of the boundary. The best observations of these diffuse streaks were performed on the second sample Sx: although the separation of satellite and lattice spots is not as large as in the Y cut slabs, satellites are in a better orientation relative to the anisotropic sample strain broadening. A striking observation is that the most intense diffuse streak exhibits a quadruple splitting, corresponding to the directions where the four satellites disappeared. In real space, these long narrow split streaks correspond to the existence of rotated planar walls, without spatial correlation: this probably corresponds to the rotated Dauphiné twin walls, observed by electron microscopy.

In the second samples (Sx) we also performed some measurements with the FRELON x-ray camera, with the larger field of view (40 mm), allowing nearly real time exposition (10 s) necessary to follow rapid variations during completion of phase transition. We then get striking evidence of the discontinuous disappearance of satellites at the inc phase boundary.

In conclusion, the high spatial resolution of RS experiments has given two major new results on the structure of the interphase region of quartz:

- discontinuous variation of satellite position at the coexistence phase boundary
- presence of a split diffuse streak, probably due to rotated planar Dauphiné twin walls.

The possibility to perform such detailed measurements was rather unexpected: the coexistence in a single quartz crystal of two phases with different lattice parameters generates large strains which in usual diffraction measurements produce a large broadening of lattice and satellites peaks. [2]. However the high resolution of the photographic plate or of the x-ray camera gives detailed topographies of lattice and satellites diffraction spots. It is then easy to correlates specific features at crystal surfaces or at rotation domains with a resolution close to that of a perfect sample. The present experimental set-up is then well adapted for detailed real time study of the quartz transition and it is possible to gain one order of magnitude in the spatial resolution.

References

[1] G. Dolino, P. Bastie, J. Phys./ Condens. Matter 13, 11485 (2001)
[2] P. Bastie, G. Dolino and B. Hamelin, J. Phys. D: Appl. Phys. 36, A139 (2003)