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IN-SITU SYNCHROTRON X-RAY TOPOGRAPHIC STUDIES OF POLYCRYSTALLINE ICE

Ian Baker and Fuping Liu

Thayer School of Engineering, Dartmouth College, Hanover, NH 03755, USA

Michael Dudley

Department of Materials Science and Engineering, State University of New York at
Stony Brook, NY 11794, USA

David Black

National Institute of Standards and Technologies, Gaithersburg, MD 20899, USA

ABSTRACT

This paper provides an overview of recent *in-situ* synchrotron x-ray topography experiments on the deformation of polycrystalline ice. It has been shown that deformation is initiated by the nucleation of semi-hexagonal basal dislocation loops at grain boundary facets. The dislocations subsequently glide across the grain. The slip system which operates was found to be the one with the highest resolved shear stress. Non-basal slip was also observed but only becomes significant if basal slip is inhibited. Whilst slip propagation between grains has been observed (once) it appears to occur only for special orientation relationships between grains.

INTRODUCTION

X-ray topography was first used to examine ice by Webb and Hayes (1967) and first applied to the study of large angle grain boundaries in ice by Hondoh and Higashi (1983). However, these and other (see Baker, 1992) early studies were compromised by the use of conventional x-ray sources, in which the x-ray intensity is such that film exposure times of the order of one hour are required. The problem is that these exposure times are long compared to the times required for dislocation recovery in ice. The result is blurred dislocation images, recovered rather than as-deformed dislocation structures and the inability to perform meaningful dynamic experiments.

A breakthrough was made by Ahmad, Ohtomo and Whitworth (1986) who used a synchrotron as a source of white x-radiation to study ice single crystals. The high intensity of this source allows photographic exposure times of only a few seconds and opened up the possibility of dynamic experiments. Only recently has synchrotron x-ray topography been used to study polycrystalline ice (Liu, Baker, Yao and Dudley,1992) and only in the last two years has this method been used to perform *in-situ* deformation studies on ice polycrystals (Liu, Baker and Dudley, 1993a).

The use of x-ray topography for the study of grain boundaries has been reviewed recently (Liu and Baker, 1994) and the *in-situ* deformation variant of this technique has been contrasted recently with the use of *in-situ* straining transmission electron microscopy (Baker and Liu, 1994). The purpose of this paper is to present an overview of recent *in-situ* straining synchrotron x-ray topographic studies of polycrystalline ice.

THE TECHNIQUE

The technique mainly used to image polycrystalline ice is the transmission Laue geometry of synchrotron white-beam x-ray topography. However, recent experiments have been performed with monochromatic radiation as discussed at the end of this section. If a white, area-filling x-ray beam impinges on a crystal, different planes select the appropriate wavelengths to satisfy Bragg's law and produce diffraction spots. The resulting Laue pattern is, typically, recorded on photographic film, see Figure 1.

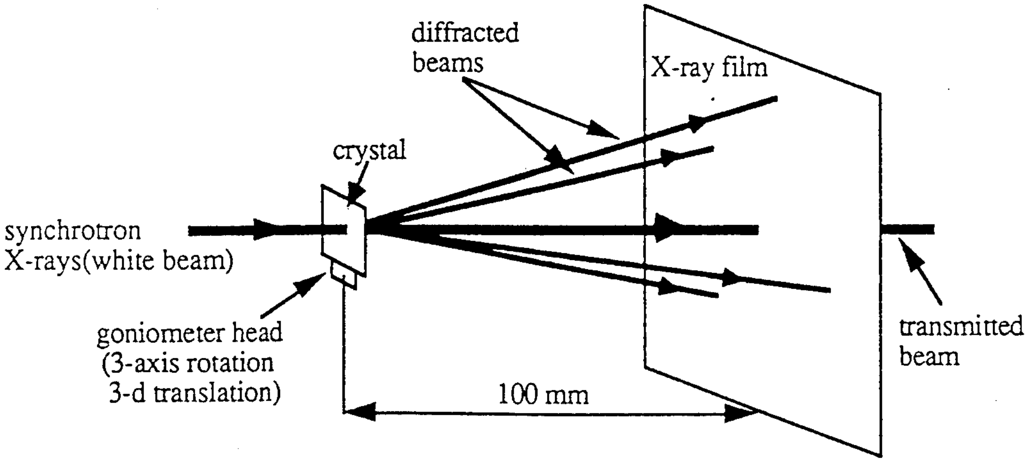

Figure 1. The arrangement of crystal, X-rays, and photographic film in the transmission Laue geometry of synchrotron-based white-beam transmission X-ray topography.

On the Laue pattern, each diffraction spot is a two-dimensional projection of the three-dimensional structure of the crystal. If the specimen is a polycrystal each grain produces a separate Laue pattern. Determination of the grain of origin of a particular diffraction is straightforward to ascertain since the shape of the diffraction spot is the same as the shape of the irradiated grain, see Figure 2. Some diffraction spots from different grains may overlap but this is easily overcome by rotating the specimen.

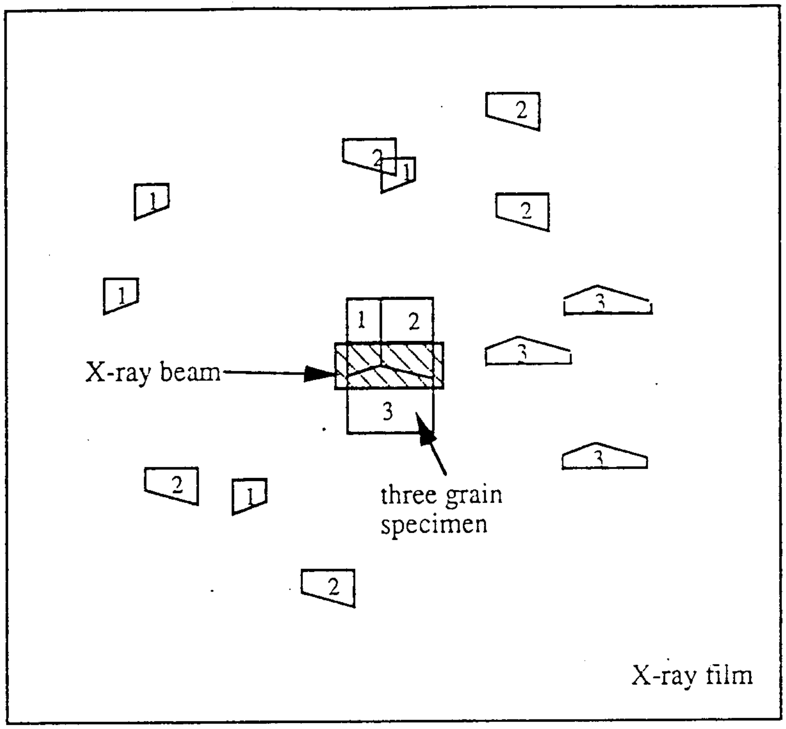

Figure 2. Three Laue patterns are formed on an X-ray film when three grains in a specimen are illuminated. From the shape of each diffraction spot its corresponding grain of origin can be identified.

Ice polycrystals up to ~2 mm can easily be studied with exposure times of one to two seconds. The disadvantages with this technique are that the best resolution is ~1-2 μm, and that the contrast of defects can be complex because more than one wavelength can contribute to each image (Tanner, 1976). Whilst the latter problem need not concern us too greatly since we are not studying the details of image contrast here, the low resolution limits both the study of dislocation dissociation into partials and the dislocation densities that can be examined. The former limitation means that it is not possible to determine whether the dislocations move on the glide planes, where they would dissociate (Gilra, 1974), or the shuffle planes, where they would not (Whitworth, 1980). The limitation on the upper dislocation density of 10^6 m^{-2} means that not only is great care required for specimen preparation and handling but also that deformation studies are limited to the regime of microplasticity. Thus, one needs to exercise caution in using the results of x-ray topography to interpret behavior at yielding and higher strains.

Even with the limitations of synchrotron x-ray topography noted above, the technique is far superior to the other techniques that have been employed to study dislocations such as etch-pitting and transmission electron microscopy (Baker, 1992). Recent results, which show that image forces can exert an influence on dislocations up to 100 µm from a free surface (Liu, Baker and Dudley, 1994) cast severe doubts on the fundamental utility of the latter two techniques for the study of ice.

As noted above, in recent studies of polycrystals, monochromatic x-radiation has been utilized. In contrast to white radiation experiments, in this method only a single diffraction spot is obtained from a precisely-oriented grain. It is improbable that two grains will be correctly oriented to produce diffraction spots simultaneously. Thus, in general, only one diffraction spot at a time is recorded from a single grain in a polycrystal. The diffracted image is viewed in real-time with an x-ray sensitive video camera so that dynamic experiments can be performed.

SPECIMEN PREPARATION

A key component of synchrotron x-ray topographic studies of ice is the specimen preparation. Typically, high-quality, high-purity, bubble-free, columnar-grained ice is grown from de-oxygenated, de-ionized water in the Ice Research Laboratory at Thayer School of Engineering, Dartmouth College, in the form of pucks which are ~300 mm high in the column direction. Specimens are cut initially with a band saw and then with a fine jeweler's saw into rectangular blocks. The crystallographic orientation of each grain is identified by the shape of etch pits (Higuchi, 1958) and the positions of grain boundaries determined by viewing specimens between crossed polarized sheets. The rectangular ice specimens are planed to ~3 mm thickness and the required area of a few cm length and width by gently shaving with a single-sided razor, taking care not to tear open the grain boundaries. The surface is carefully removed by polishing it with a smooth cloth soaked in ethyl alcohol until its thickness is ~2 mm. The alcohol adhering to the specimen surface is removed by a wash in a n-hexane bath. The final specimens, measuring typically 25 mm X15 mm X2 mm, are mounted on a special polymer base, which has the same linear thermal expansion coefficient as ice, and annealed at -10°C for 15 days. For examination, the specimens are cooled to -20°C very slowly, and transported to the National Synchrotron Light Source, Brookhaven National Laboratory (BNL) on Long Island, NY. *In-situ* deformation studies are performed using a specially-built compression jig assembled with a cryostat (Liu and Baker, 1993) and the National Synchrotron Light Source at BNL. Throughout, care has to be taken to avoid subjecting the specimens to thermal shock which will result in the formation of numerous dislocation loops (Liu, Baker, Yao and Dudley, 1992), see Figure 3.

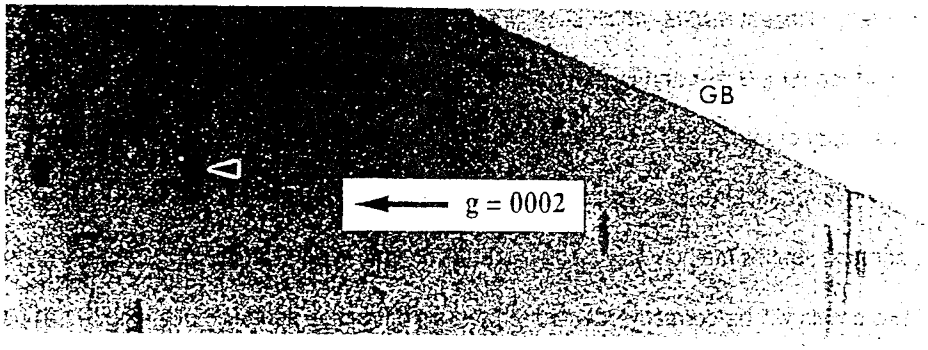

Figure 3. Dislocation loops formed in ice after subjected the specimen to a high cooling rate.

GRAIN BOUNDARY NUCLEATION OF BASAL DISLOCATIONS

In the numerous topographs obtained from polycrystalline ice there are two dominant features. First, dislocation nucleation occurs predominantly at the grain boundaries. Second, the dislocations which are nucleated are semi-hexagonal $<11\bar{2}0>$ loops with their sides in screw and 60° orientations which lie on the basal plane (Liu, Baker and Dudley, 1993), see Figure 4.

Although grain boundary nucleation overwhelms any lattice or surface nucleation of dislocations, a question raised by initial observations of grain boundary nucleation was whether nucleation occurred at the grain boundary interior or at the line of intersection of the grain boundary with the free surface since stress concentration are quite evident there. More recent observations have shown that whilst the latter process probably does occur, nucleation from the grain boundary interiors is common, see Figure 5.

Earlier, Whitworth and co-workers (Ahmad, Ohtomo and Whitworth, 1986; Ahmad and Whitworth, 1988, Shearwood and Whitworth, 1991; Shearwood and Whitworth, 1992) observed the nucleation of dislocations either within the lattice or at the specimen surface at scratches, in ice single crystals. However, neither of these events appear to be important in polycrystalline ice presumably because the internal local stress is much higher than the external stress. The stress-concentrating effect of grain boundaries is evident as a darkening of the image there. This darkening has often been observed to be reduced after dislocation emission, indicating that the stresses are plastically relaxed, see Figure 6.

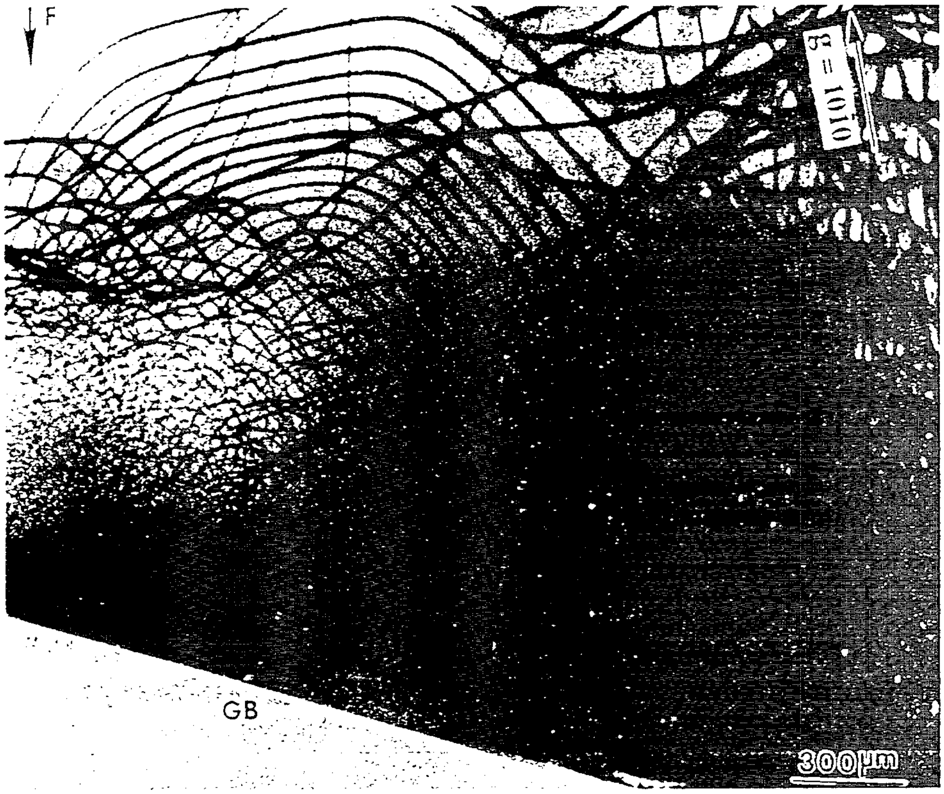

Figure 4. A monochromatic x-ray topograph showing a large number of basal semi-hexagonal dislocations generated at the grain boundary after a load of 0.3 MPa for 20 minutes at -16.3°C.

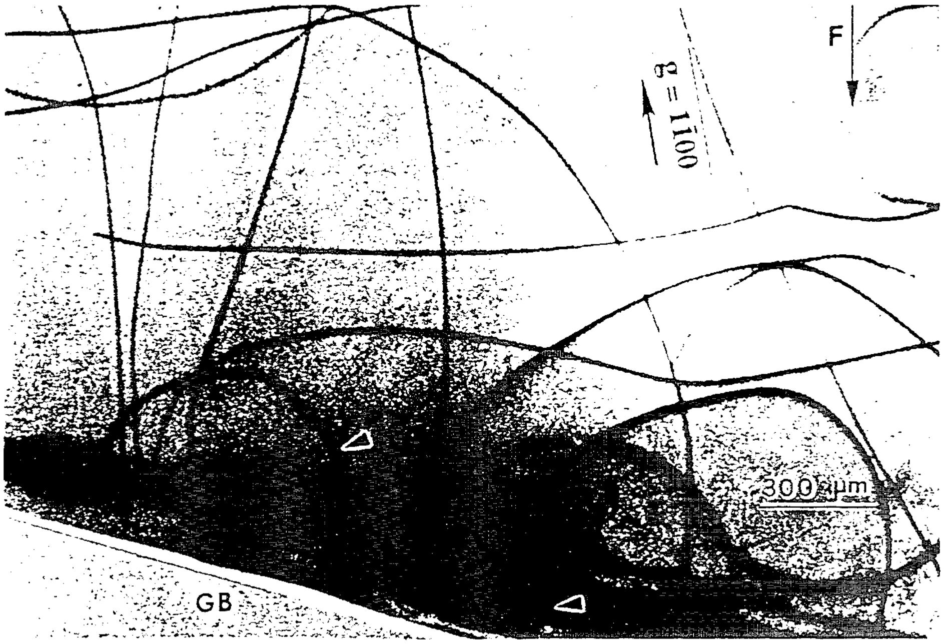

Figure 5. A monochromatic x-ray topograph showing the microstructure after a vertical compressive load of 0.1 MPa for 30 minutes at -16.3°C followed by an anneal at -10°C for 5 days.

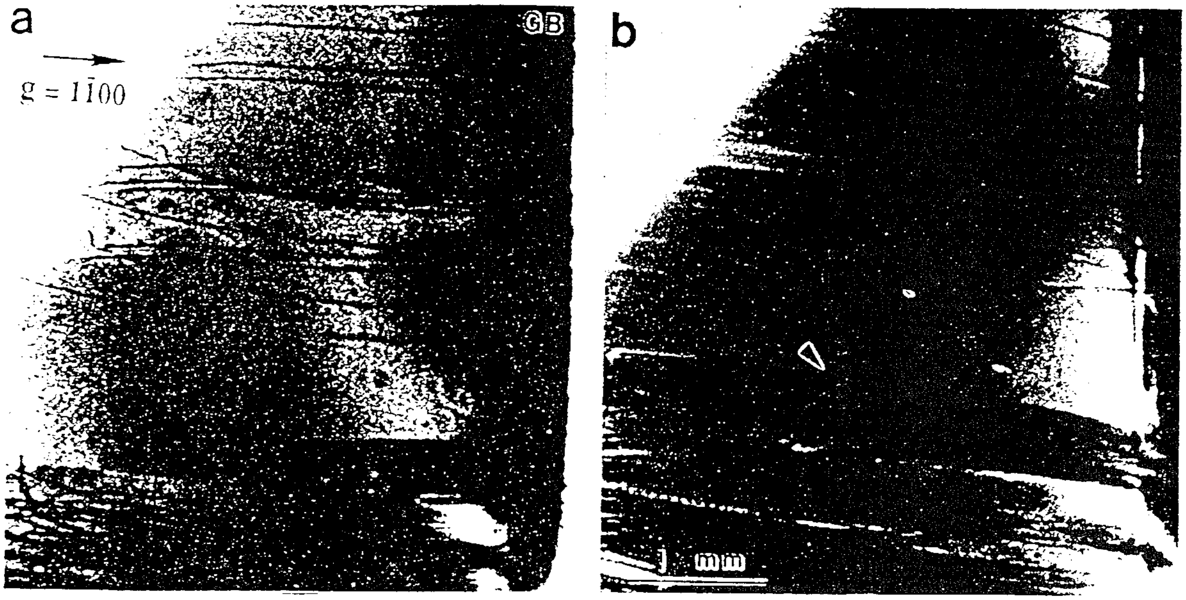

Figure 6. Two white beam X-ray topographs showing that the darkening at the grain boundary in (a) was reduced at -15°C after dislocation emission (b) , indicating that the stresses are plastically relaxed.

The basal slip system with the highest Schmid factor was found to be the most active in polycrystalline ice whereas the grain boundary orientation relative to the loading direction seemed relatively unimportant (Liu et al., 1994a). The role of the misorientation between two grains is to enhance dislocation generation at grain boundaries through elastic incompatibility and to prevent dislocation transmission through grain boundaries through plastic incompatibility (Liu et al., 1994a). Dislocations are nucleated at facets at the grain boundaries. Figure 7 shows dislocations which have been nucleated at facets and subsequently traversed both grains.

OTHER FEATURES

There are a number of other features that have been observed on x-ray topographs of less importance. If the dislocations in a grain are generated at the grain boundaries, one question that arises is whether the dislocations nucleate at the grain boundary or whether slip propagates from the adjacent grains. Thus far, only a single observation of slip propagation from one grain to another has been observed (Liu, Baker and Dudley, 1994a). In that case, the two grains had quite a special orientation relationship. Basal dislocations nucleated at a grain boundary in one of the grains traversed the grain and entered the opposite grain boundary. These stress concentrating effect of these dislocations presumably eventually led to the basal slip that was observed in the adjacent grain.

Dislocation pile-up formation has been observed at grain boundaries (Liu, Baker and Dudley, 1994b). However, that these pile-ups cause dislocation nucleation in the adjacent grain has not yet been observed.

A notable feature of all the above topographs is that dislocations are gliding predominantly on the basal plane. What happens if the resolved shear stress for basal slip is zero? In that case non-basal edge dislocation segments are nucleated at the grain boundaries and move rapidly across the grain but the screw segments that they trail behind them appear to be immobile (Liu, Baker and Dudley, 1994b). (It has not been possible to determine on which plane the short edge segments glide.) Thus, the non-basal dislocations can not contribute much to the plastic strain. Interestingly, even when the resolved shear stress for slip on the basal plane from the far-field stress is close to zero, semi-hexagonal basal dislocation loops can still been nucleated at the grain boundary (Liu, Baker and Dudley, 1994b). These loops presumably arise because the local stress state at the grain boundary is different from the stress state imposed by the far-field stress. However, these dislocations do not glide far into the grain since the resolved shear stress presumably falls rapidly to zero.

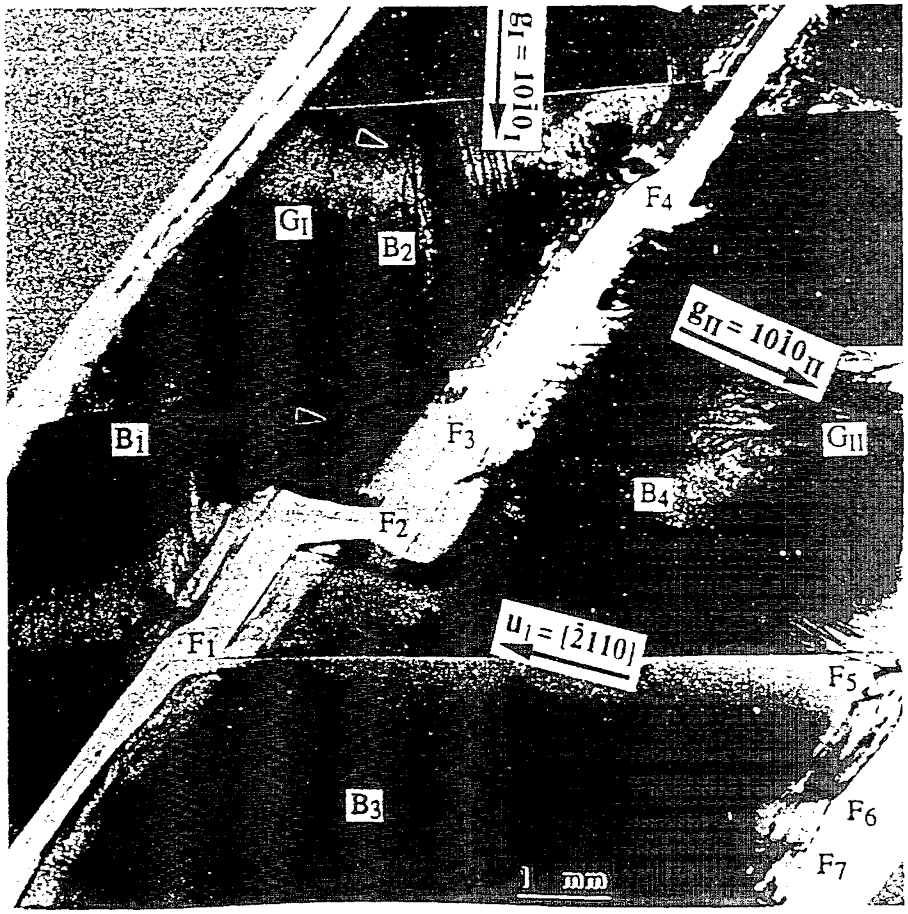

Figure 7. A composite X-ray topograph showing dislocation structures in two adjacent grains at -10°C.

CONCLUSIONS

In-situ deformation experiments on polycrystalline ice using synchrotron x-ray topography have shown that deformation proceeds by the nucleation of semi-hexagonal basal dislocation loops at grain boundary facets. These dislocations, once nucleated glide across grains and in some cases pile up at the opposite grain boundary. The slip system with the highest critical resolved shear stress has been found to be the one that operates. Non-basal slip can occur but only becomes significant if basal slip is inhibited. Thus far, slip propagation between grains has been observed only once and then only for grains for which a special orientation relationship exists.

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