Faceting and Roughening of the Asymmetric Twin Grain Boundaries in Zinc

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Faceting of grain boundaries (GBs) or surfaces can be considered as a phase transition when the original surface or GB dissociates onto flat segments whose energy is less than that of original surface or GB. Zn $[11\bar{2}0]$ flat single crystals were grown using the modified Bridgman technique from Zn of 99.999 wt. % purity. Individual elongated twin plates having very uniform thickness were produced with the aid of slight deformation of single crystals. Parallel elongated sides of the twin plate are formed by the coherent symmetric twin $(1\bar{1}0\bar{2})_1 \| (1\bar{1}0\bar{2})_2$ grain boundary (STGB) facets. Due to its optical anisotropy, zinc allows one to study the shape of the GB with the aid of polarised light. The stationary shape of the slowly migrating tip of the twin plate has been studied *in situ*. The hot stage of optical microscope was used. The temperature interval from 592 to 692 K was investigated. Below 632 K the twin tip contains only one plane facet *1* which is nearly parallel to the $(\bar{1}10\bar{2})_2$ plane and has the angle of 84° with the coherent STGB. Above 632 K the second facet *2* appears at the tip of the twin plate. This facet is nearly parallel to the $(1\bar{1}00)_1$ plane and has the angle of 46° with the coherent STGB. Between 632 and 682 K both 84° and 46° facets coexist, and 84° facet

gradually disappear with increasing temperature. Above 682 K only 46° facet is present in the twin tip. The indications of the GB roughening phase transition were also observed, namely the edges of the facets become smoother with increasing temperature. The GB phase diagram for the twin GBs in zinc containing the lines of two GB faceting phase transitions has been constructed. Schematic Wulff-Herring diagrams explaining these transitions are presented.

Keywords: grain boundaries, faceting, roughening, Zn, twins, phase diagrams

1. Introduction

Faceting of grain boundaries (GBs) or surfaces can be considered as a phase transition when the original surface or GB dissociates onto flat segments whose energy is less than that of original surface or GB [1]. Recently, the faceting of vicinal free surfaces became a subject of intensive investigations [2–4]. One of the reasons is that the faceted surfaces contain a very regular equilibrated one-dimensional grid of steps or two-dimensional array of pyramids. Such faceted surfaces can be used as the substrates, respectively, for quantum wires or quantum dots opening the way to the nanoelectronics. The method of phase diagrams describing ordering, faceting and roughening of free surfaces can be applied also for GBs. It is important to study *in situ* the GB faceting and especially roughening in order to observe the GB structure at elevated temperature [5]. Zn was chosen for this work since it has non-cubic lattice and possesses the optical anisotropy. Therefore, Zn offers a possibility to study the GB shape in polarised light directly in hot-stage of the optical microscope. This method was originally developed for the investigations of GB migration [6–10]. The disadvantage of the Zn non-cubic lattice is that the ratio of *a* and *c* spacing is not rational, and the simple coincidence site lattice (CSL) cannot be constructed for Zn GBs. The approach of constrained CSLs (CCLSs) has to be exploited [11, 12]. Therefore, the most simple case of twin GBs was chosen for our first *in situ* investigation of GB faceting and roughening in Zn. The Σ3 twins in cubic metals were proved as very good objects for the investigation of faceting. For example, different Σ3 CSL facets like $(010)_{CSL}$ and $(110)_{CSL}$ and off-CSL 82° 9*R* facet were observed at the end of Cu twin plates in different conditions [13–16].

2. Experimental

Zn $\left[11\bar{2}0\right]$ flat single crystals were grown using the modified Bridgman technique from Zn of 99.999 wt. % purity [6–8]. Individual elongated twin plates having very uniform thickness were produced with the aid of slight deformation of single crystals. The produced twin plates are perpendicular to the surface of the sample and possess a very uniform thickness. The $\left[11\bar{2}0\right]$ axes in both grains are also perpendicular to the surface of the sample. The parallel elongated sides of the twin plate are formed by the coherent symmetric twin $(1\bar{1}0\bar{2})_1||(1\bar{1}0\bar{2})_2$ grain boundaries (STGBs) Due to its optical anisotropy, zinc allows one to study the shape of the GB with the aid of polarised light. The stationary shape of the slowly migrating tip of the twin plate has been studied *in situ* in the hot stage of optical microscope in the temperature interval from 592 to 692 K. The samples were protected from oxidation by pure nitrogen atmosphere. Before measurements the samples were electropolished in the H_3PO_4+C_2H_5OH solution. An additional polarisation filter applied in the reflected beam permits to distinguish different orientations of the grains by the different intensity of the reflected light. GB shape was recorded in the course of experiment by colour video camera connected with a microscope and video recorder. This method was originally developed for the investigations of GB migration [6–8]. The driving force resulted from the GB phase transitions is usually rather low to change the GB shape in a reasonable time. Therefore, we use the constant capillary driving force to induce the GB migration. During the slow movement forced by capillarity the GB has the opportunity to reach the equilibrium shape which corresponds to the temperature of the experiment. This method was used, for example, in our studies of the "special GB – general GB" phase transitions [17, 18].

3. Results and discussion

The micrographs in Fig. 1 show the change of the shape of GB at the tip of the twin plates in Zn with increasing temperature. The shape of the twin tip differs drastically from the rounded shape of the GB half-loops in Zn bicrystals containing non-special GBs [7–10]. At low temperature the twin tip contains one flat facet *1* lying almost perpendicular to the STGBs (Fig 1a). With increasing temperature a second facet *2* appears at the tip (Fig 1b). The facet *2* has an angle of approximately 45° both with STGB and the facet *1*. When temperature increases further, the length of the facet *2* also increases and that of the facet *1* decreases (Fig. 1c). Above certain temperature only the facet *2* is present at the twin tip having rather sharp edges with STGBs (Fig. 1d). Close to the Zn melting temperature T_m the edges at the intersection of the facets become rounded, but the flat part of the facet *2* at the tip is still visible (Fig. 1e). In Fig. 2 the temperature dependence of the angles between STGB and the facets *1* and *2* is shown. The mean value of the angle for the facet *1* is 84° and for the facet *2* is 46°. Below 632 K only facet *1* is present in the twin tip. Between 632 K and 677 K the facets *1* and *2* coexist. Above 677 K only facet *1* is present. Schemes in Fig. 3 demonstrate the CCSLs and crystallography of the observed facets. STGB is coherent and coincide with $(1\bar{1}0\bar{2})$ planes in both lattices (Fig. 3a). Facet *1* is nearly parallel to the $(\bar{1}10\bar{2})_2$ plane, and the coincidence is much worser that in case of the coherent STGB (Fig. 3b). Facet *2* is nearly parallel to the $(1\bar{1}00)_1$ plane (Fig. 3c). In Fig. 4 the temperature dependence of the normalized lengths a_1 and a_2 of the facets is presented. a_1 decreases monotonically from 1 at 607 K to 0 at 682 K. a_2 increases from 0 at 607 K reaching the maximum of 0.95 at 682 K and decreases to 0.58 at 692 K. The decrease of a_2 the proceeds in the absence of other facets. Above 682 K the form of the tip becomes rounded. The disappearance of the sharp edges at the intersection of GB facets can be explained by the roughening of the GB facets close to the melting point. In Fig. 5 the phase diagram for the faceting of twin GBs in Zn is constructed. Two GB faceting phase transitions proceed: (1) Between 677 and 682 K the facet *1* disappears. (2) Between 607 and 632 K the facet *2* appears. STGB is stable at all temperatures. In Fig. 6 the schematic Wulff-Herring plots are constructed illustrating the possible energetic reasons for both faceting GB transitions and roughening of the facet *2*. The energetic minimum for the STGB is present at all temperatures. At 600 K the minima for both *1* and *2* tip facets exist, but the facet 2 is metastable. At 650 K both facets *1* and *2* are stable and present in the equlibrium form. At 680 K the roughening starts, the energetic minimum for the facet 1 disappears, and only facet *2* is stable and present in the equlibrium form. At 690 K the roughening continues, the energetic minima for both remaining facets become schallower and the rounded part of GB loop progressively consumes flat facets.

The *in situ* observation of the GB shape allowed us to observe for the first time the GB faceting phase transition when one smooth facet exchange the another one. Previously only two kinds of GB faceting phase transition phenomena were onserved: (1) Reversible GB roughening with increasing temperature and dissociation of smooth roughened GB into facets at decreasing temperature in Al and Au [5]. (2) Dissociation of the originally flat GB onto flat facets whose energy is less than that of original GB [13, 14]. GB facets can be observed only close to the coincidence misorientations. In [17, 18, 19] it has been shown that the GB possess the special structure and properties in the limited areas of temperature T and misorientation θ close to a coincidence misorientation θ_Σ. In other words, by increase of $\Delta\theta = |\theta - \theta_\Sigma|$ and T the phase transition "special GB – general GB" proceeds and GB looses its special structure and properties, particularly, the GB facets disappear. The ratio of *a* and *c* spacings in Zn is not rational and is temperature dependent. Due to this fact the direct transitions "special GB Σ_a– special GB Σ_b" by changing of T and θ are possible in Zn [11, 12]. Therefore, the unusual 84°–46° GB faceting transition can be driven by the transition from special GB Σ15a to another special GB Σ28a, or from Σ28a to Σ13a with increasing temperature [11, 12].

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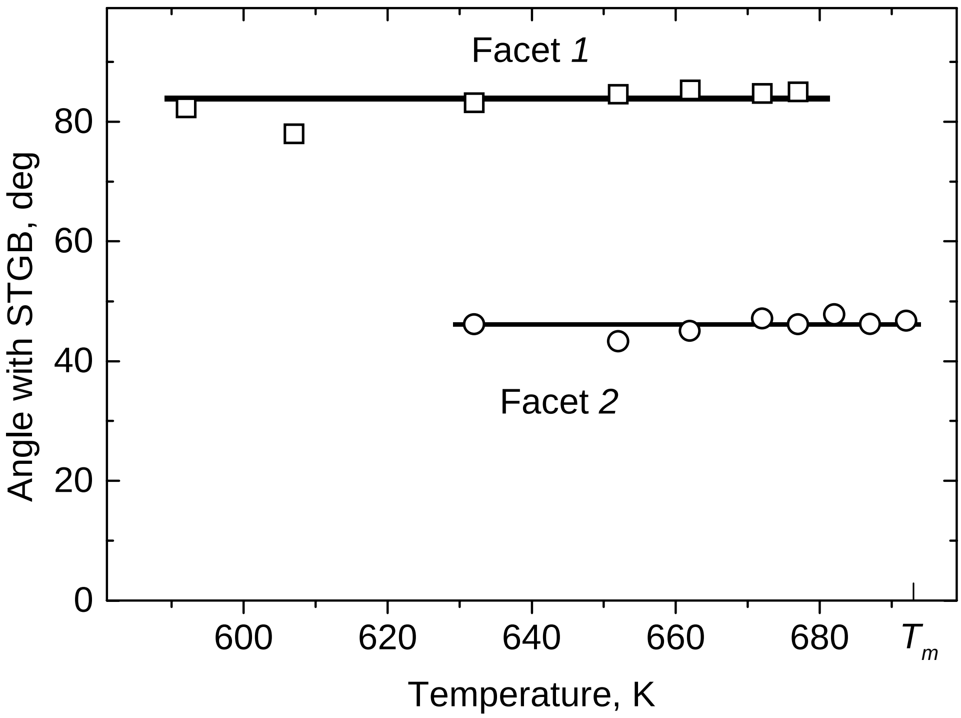

Fig. 2. Angle between STGB and the facets forming the end of twin plates in Zn $\left[11\bar{2}0\right]$ flat single crystals at various temperatures. Circles denote the $(110)_{CSL}$ facet and squares denote the $(010)_{CSL}$ facets of the twin plates.

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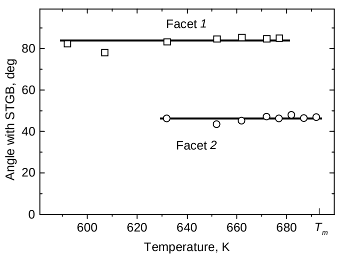

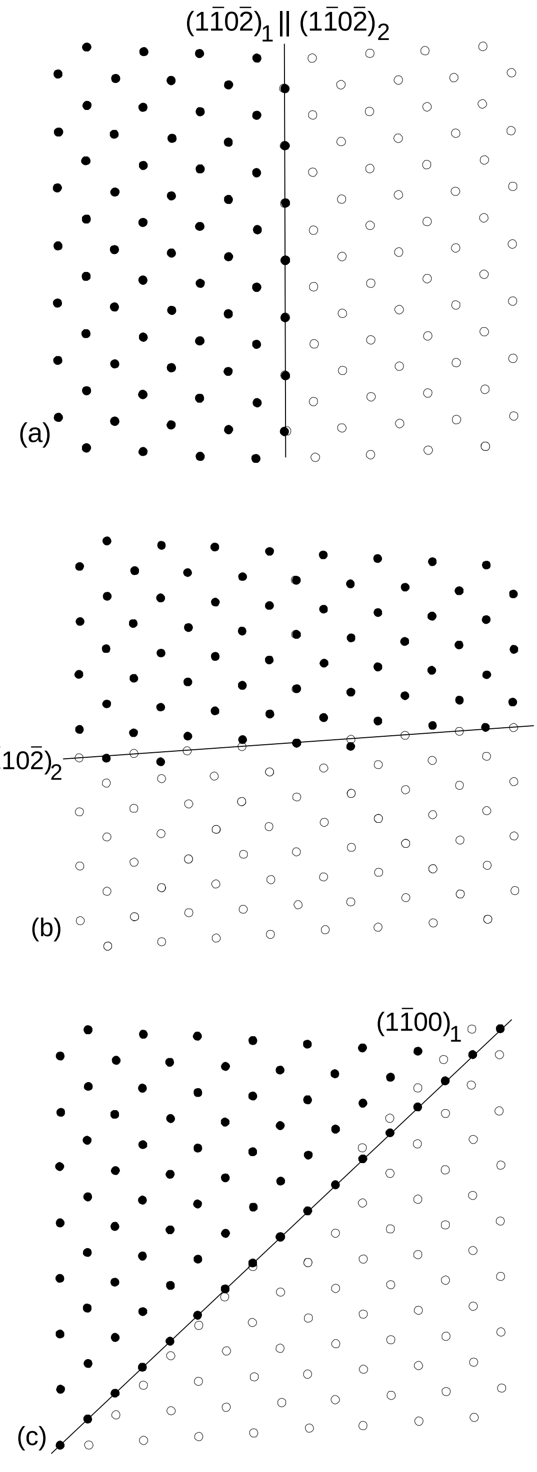

Fig. 3. Crystallographic features of the observed facets. (a) STGB $(1\bar{1}0\bar{2})_1 \| (1\bar{1}0\bar{2})_2$. (b) Facet *1* is nearly parallel to the $(\bar{1}10\bar{2})_2$ plane and has the angle of 84° with the STGB. (c) Facet *2* is nearly parallel to the $(1\bar{1}00)_1$ plane and has the angle of 46° with STGB.

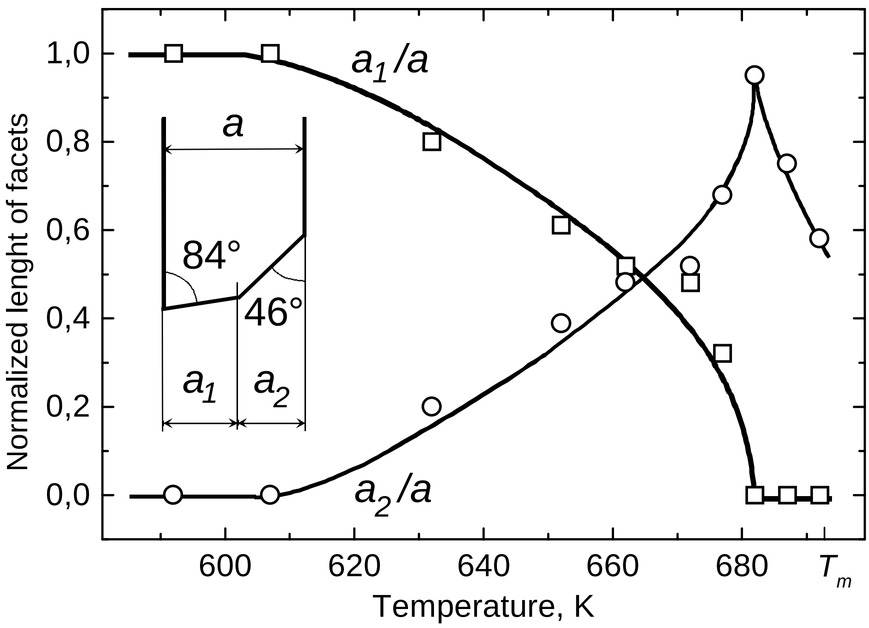

Fig. 4. Temperature dependence of the normalised length of the facet *1* (84°, a_1/a, squares) and facet *2* (46°, a_2/a, circles) of the twin plates in Zn $[11\bar{2}0]$ flat single crystals.

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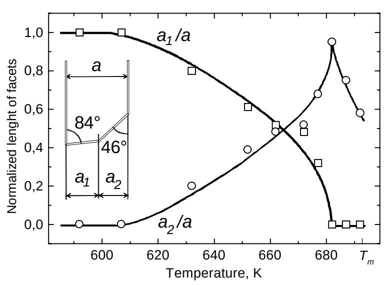

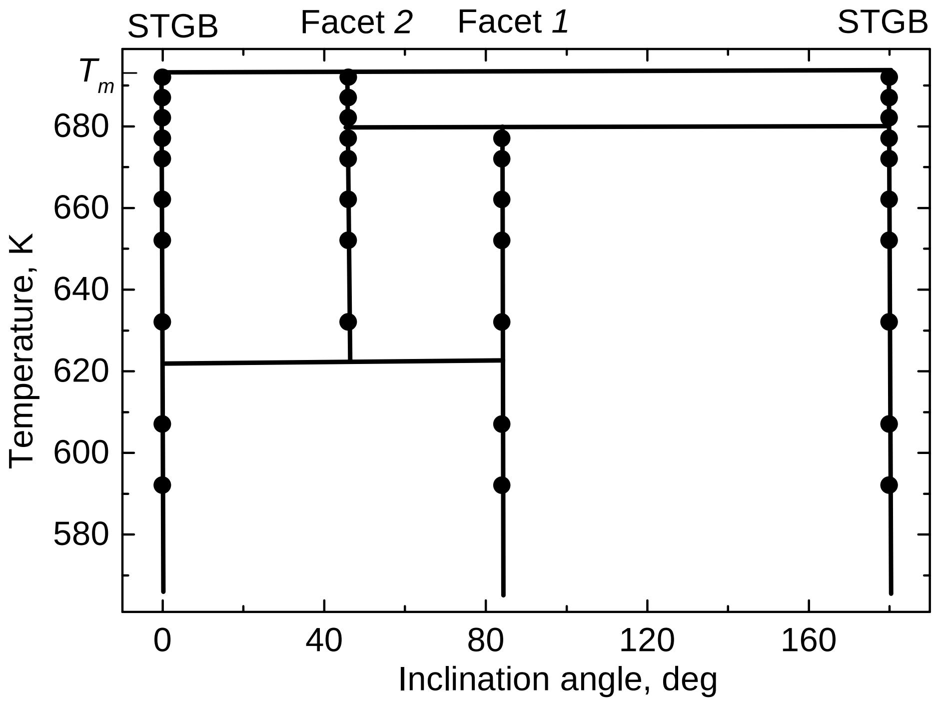

Fig. 5. Phase diagram for the various Zn twin facets. T_m is the melting temperature. The inclinations of facets are shown at the top of the diagram. Circles denote the experimentally observed facets. Vertical lines denote the temperature ranges of the stability for the GB facets. Horizontal lines denote the upper and lower limits of stability for the facet *1* (84°) and the facet *2* (46°).

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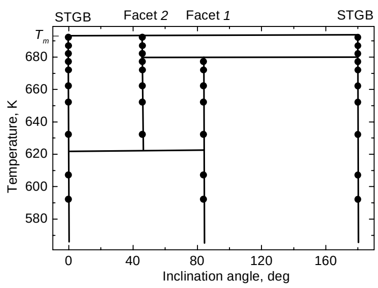

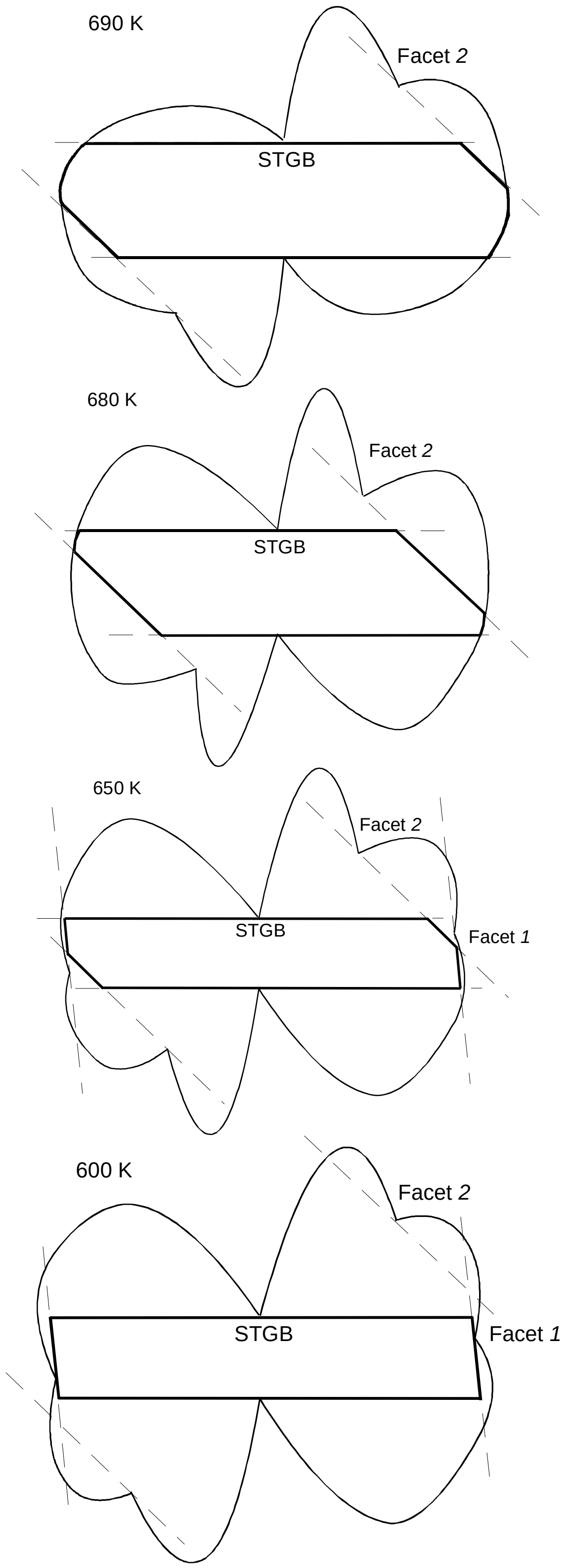

Fig. 6. Schematic Wulff-Herring plot for the twin plates in Zn $\left[11\bar{2}0\right]$ flat single crystals at various temperatures.

Figure captions

Fig. 1. Shape of the twin plates in Zn $[11\bar{2}0]$ flat single crystals at various temperatures. (a) 632 K; (b) 672 K; (c) 677 K; (d) 682 K; 692 K. Length of the scale bar is 100 μm.

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