Decoding Pattern Dependence of Micro-indentation on Crystal Orientation of Cube-textured Aluminum Foil Using SAXS and Temperature Changes

S. Saimoto¹, M. A. Singh², M. R. Langille¹ and C. Gabryel¹

¹ Mechanical and Materials Engineering, Queen’s University, Kingston, ON, Canada, K7L3N6
² Physics, Eng. Physics & Astronomy, Queen’s University, Kingston, ON Canada, K7L3N6

Abstract

Recent use of nano-indentation to assess work-hardening of complex microstructure of dual phase steels as a function of deformation together with crystal plasticity finite element method to simulate the bulk properties gives rise to the question of the role of crystallite orientation during indentation. Such studies using nano-indentation on µm-sized grains are difficult. To elucidate this effect, Vickers micro-indentation of cube textured Al capacitor foils were examined. These foils of 106 µm thickness are convenient for small angle X-ray scattering (SAXS) examination and the orientation around the indent can be examined by back-scattered electron diffraction (EBSD). The indentations were carried out with the diagonals parallel to the rolling direction (RD) and 45° to it at temperatures from 20 °C (295 K) to 160°C (433 K). The SAXS tests were performed only at 20°C on RD specimens.

Keywords: Aluminum, micro-hardness, SAXS, stacking fault tetrahedral, dislocation intersections

1. Introduction

Indentation testing using Vickers hardness with diamond pyramid head has been the standby materials characterization of mechanical properties. Since the discovery of age-hardenable alloys, the great progress is due to the advent of hardness testing to monitor ageing as function of time and annealing temperatures and has become the standard procedure to perform such tests to delineate the optimum hardness for alloy development. As the microstructure of the alloy becomes more complex with multiple hardening precipitates, the indenter size have decreased to micro- and nano-meter scales. Detailed characterization of texture and stored work in damaged volumes using conical indenter have been performed to determine if a simple correlation between hardness and geometric-necessary-dislocation (GND) density exist [1]. The application of such computational analyses was recently illustrated for dual-phase steels [2] in which nano-indentation result was used to correlate to the bulk stress-strain response. However application of such methods to face-centred cubic (fcc) structures may require more statistical testing to assess the crystal orientation dependence of the hardness response. Hardness has been attributed to two main factors; the yield strength due to the size, density and distribution of the embedded nano-particles and the gradients in GND densities around the indent interface. Brown [3] has simulated indentation using Vickers diamond indenter and has developed a circular flow model to predict lattice rotation which could rise as high as 12°. Such rotations have been observed in Berkovich-tip indented matrix of cube oriented copper crystal using Kikuchi line analysis of electron diffraction (TEM) images (14°) [4] and in conical indented (111) surface of copper crystal (15°) using EBSD [1]. Although the analytical means for modelling micro- and nano-plastic flow seems to be at hand [2], for aluminum (Al) alloys dynamic recovery can occur during indentation at room temperatures.

During the recent work of assessing double Bragg scattering (DBS) giving rise to streaks [5] when performing SAXS of Al alloys, it was convenient to use cube textured Al foils to show that crystal orientations exist which do not satisfy the DBS diffraction conditions. The occurrence of such streaks need to be removed prior to the analyses of isotropic data to reveal nano-void formation. During this study it was discovered that the orientation of the indent diagonal whether it was parallel to the rolling
direction (RD) <100>, or at 45° to it, <110>, the shape of the indent was noticeably different. The origin of this indent pattern difference is examined in this report as a function of temperature.

2. Background on slip systems and interactions in crystals of multiple slip orientation

2.1 Multiple slip effects in copper single crystals under homogeneous deformation

Many past studies [6 - 11] have shown that shape-change of high symmetry oriented crystals do not necessarily occur according to geometric considerations of resolved shear stress since the strengths of dislocation interactions are not all equal. Furthermore, two different slip systems which occur in different parts of the crystal may meet to form strain-compatible dislocation boundaries [7]. One case is shown in Fig. 1, whereby slip traces on (011) surface of [011] oriented tensile axis pulled at 78 K [6] correspond to primary slip on one side and critical slip on the other to form {001} boundaries as predicted by Kear [7]. As the strain increase, the two portions rotate in opposite directions resulting in break-up of the crystal. The cause of this observation is due to the instability of {011} under tension. However even under stable compression testing using temperatures ranging from 78 K to 813 K, it was found that the predicted four slip systems reduced to two above 295 K [8]. The pair combination may give rise to either Lomer-Cottrell (primary-conjugate) or Stroh-Hirth (primary-critical) locks. The observation indicate that at low temperatures the sessile jogs form from primary-critical intersections to produce vacancies by jog-dragging but at elevated temperatures like-jogs combine to become sufficiently long to dissociate into sessile Stroh-Hirth locks such that strains from this combination ceases. The formation geometry of Lomer-Cottrell locks are more restrictive and they can also be unzipped such that the primary-conjugate combination can continue to operate to accommodate the imposed strain. Strain rate sensitivity using thermal cycling between 78-162 K shows that [001] which includes both primary-critical and the primary-conjugate pairs. On the other hand for the 78-273 K cycling, the response are identical showing that the primary-critical obstacle formation, that is jog-dragging, has ceased due to formation of Stroh-Hirth locks [9] as indicated above.

2.2 Multiple slip effects in aluminum single crystals

During tensile deformation of [001] Al single crystals, it was found that serrated flow initiated at about 250 K and the critical strain decreased with increasing temperature. These serrations coincided with the appearance of slip lines [9] on the (110) surface wherein three traces are observed with the third and fourth being coincidental. This instability was attributed to strain ageing due to atmosphere formation by vacancies. Since the volume fraction of cube texture in Al sheets has a large effect on its formability, basic studies have been undertaken on plane strain compression of (001) [110] and (001) [010] single
crystals at various temperatures [10, 11]. It was found that above strain of 0.20, break-up of the crystals according to the primary-critical intersections were observed with formation of {001} deformation boundaries. Furthermore the increase of cube texture in sheet occurs during recrystallization of rolled sheet wherein the recrystallization nuclei are found in the high GND regions due to strain gradients. To examine this effect, the occurrence of recrystallized grains after Vickers indentation of 12 % pre-rolled sheet of nominally pure aluminum of mm-sized grains were assessed as function of orientation by Xu et al. [12]. For the (311) [215] orientation, the indent shape had bulged out; that is sides were convex indicating that the crystal along the indent interface was pushing up along the indenter faces from the original surface plane. These indented grains which were annealed at 310 °C for 1 h showed that grains with (001) parallel to the sheet surface did not form new crystallites whereas most other orientations showed various degrees of recrystallization. This observation is in accordance with that for compression of Al single crystals, in which Nakata [13] found that after about 10 % compression of [001] crystal the amount of stored work was about 5% compared to 25 % for all other orientations including polycrystalline specimen. It should be kept in mind that the primary-critical intersections results in sessile jogs which generate vacancies that would promote dynamic recovery. These aspects of deformation behaviour will be discussed in the slip pattern changes caused by pyramid indenter on (001) with diagonal in RD and 45° orientations at various temperatures.

3. Experimental details
High purity aluminum capacitor foils supplied by Toyo Aluminum of Japan were 106 µm thick sheets that were fully recrystallized with 89.9 % cube texture. Specimens about 25 x 20 mm were bright polished with etch solution of 10 g NaNO₃, 100 ml saturated NaOH and 80 ml distilled H₂O. After rinsing in dilute HCl to remove sodium on the surface, electro-polishing was done using in volume % solution of 7 % perchloric acid, 70 % ethanol, 20 % distilled water with balance butyl-cellusolve; the current density was 1.5 A/cm². Pre-cooling of apparatus and solution, permitted polishing below -10°C. Vickers micro-hardness indents were made with the rolling direction (RD) either parallel to the diagonal (D) of the indenter, [100] or at 45° to it, [110]. For the elevated temperature tests, a programmable hot plate with X-Y control was used as described elsewhere [14] and the top surface was checked by a portable infra-red thermometer. From the indent geometry, not only the diagonals D but also the minimum width W were determined such that D/W can be used as a characterizing parameter with temperature change. For SAXS examination, only the RD type indents were used and the approximate D = 100 µm indents were placed about 300 µm apart. X-ray beam was collimated with two right angle slits set at 600 µm apart such that the spot size was near 1mm x 1 mm to simultaneously irradiate at least 4 indents. The details of the SAXS measurements are described elsewhere [5] but for the current purpose it is pointed out that the 3x3-hole specimen holder could be rotated about a horizontal axis parallel to RD. The rotation up to 12° were performed remotely over the same volume. In this manner the probable source of DBS could be elucidated.

4. Results: Observations of surface upheaval and rotated structures around the indent
The slip pattern around the Vickers indentation are shown in Fig. 2. The indent with the diagonal parallel to the [T00] (referred as RD) shows single slip trace parallel to the interface and D/W is 1.53 whereas for D parallel to [T10] (referred as 45°) careful scrutiny indicate two fine slip traces, nearly 90° to each other with D/W = 1.73. These fine traces become undetectable near the minimum W location near the centre that correspond to {001} dislocation boundaries described in Fig. 1. Since for isotropic deformation, D/W is 1.414, the calculations shows that the interfaces parallel to RD (the 45° one), that is the {001} boundaries, is more resistant to deformation than that at 45° to RD. Examination of the standard [001] stereographic projection (not shown) indicate that for RD orientation the traces are due to (111) on one side such that for equal operation of collinear slip the effective Burgers vector (BV) is
[1\overline{2}]\); on the other side for (1\overline{1}1), BV is [\overline{1}12]. On the other hand for the 45° case, the combination set of (\overline{1}11) and (1\overline{1}1) which upon equal strain on each, BV effectively becomes [0\overline{1}1] and for the set of (1\overline{1}1) and (111), [\overline{1}0\overline{1}]. Thus, possible wavy slip may be replaced by \{0\overline{1}1\} <\overline{T}01> slip [15] as indicated in Fig. 3. Note the Schmid factor for \{0\overline{1}1\} slip is larger than that for \{1\overline{1}1\} at [001].

![Figure 2](image-url) **Figure 2.** The indents formed at 295 K due to D parallel to RD (left) and 45° (right) are shown. Note the slip lines for RD are parallel to the trace of the interface whereas for the 45° one it is at 45° to it.

![Figure 3](image-url) **Figure 3.** This indent occurred on part of sub-boundary such that the fourth interface region (lower right side) shows slip traces parallel to \{0\overline{1}1\}. EBSD pattern [5] shows that the sub-boundary cannot be rotated by 45° to form \{1\overline{1}1\} traces. Line markers are 100 μm.

Figure 4a shows the length measurements of D and minimum W and their ratios at various test temperatures for the 45° orientation. The slope of the D line is 0.283 and is larger than that for W at 0.131. However D/W is constant with temperature. These results indicate that the work-hardening mechanisms are temperature dependent but remain the same. On the other hand in Fig. 4b for RD orientation, D is linear with temperature with slope of 0.278 but that for W shows a break near 80°C (353 K) wherein the temperature at which annealing of stacking fault tetrahedral (SFT) occurs and ends near 120°C (393 K) [16] as denoted. The change of mechanism is attributed to the termination of SFT formation leading to primary-critical jog migration and their increase in length to form Stroh-Hirth locks. The work-hardening change is indicated by W becoming temperature independent.

In order to bypass the difficulty of quantifying the evolution of the indent using the observed slip traces, the Brown mechanism of circular flow [3] was invoked. In this model, lattice rotations of about 12° are predicted about the axis normal to the plane of V-shape indent. The circular flow are centred at the points of contact of the indenter interface to the specimen surface with the radius equal to W; that is, one semi-circle intersects the other at the mid-point below the V-shape indentation (Fig. 2 in [3]). The maximum lattice rotation which is independent of indent depth is found near the edges of the interfaces. In order to validate the crystal rotation, recent work [5] using SAXS indicated that DBS should occur if one of the possible diffraction poles coincided with (90° - \theta_{Bragg}), a continuous circle
within the stereographic projection. Thus SAXS images were taken with the rotation axis of the specimen holder parallel to the RD for the RD-specimen. The specimen was solid-body rotated up to +/- 12° to detect possible streaking. However, intrinsic to the Brown model, lattice rotation occurs to result in an asterism effect up to 12°. Thus for the RD specimen the possible axes are [1\(\bar{1}0\)] and [10\(\bar{1}\)]. Subsequent to this deformation-induced lattice rotation, solid-body rotation up to +/- 12° were performed. Figure 5 schematically displays the observed streaks at the +12° position. Quantitative analysis using \{420\} and \{422\} pole figures indicate that the streaks can arise due to DBS from these planes. The details are not shown due to space limitations. The pole figures from EBSD determination [5] show that the cube texture is not ideally orientated and consists of two major closely-related orientations which are off cube axes by about 5°. For this reason, rotation about -12° did not show streaks; that is, the expected symmetry for ideal cube was not observed.

![Cube-Texture of Al Capacitor Foil](image)

**Figure 4.** The parameter, length of D and W, and the ratio of these lengths versus temperature.
(a) For 45° indents the slope for D and W is constant.
(b) For RD, only that for D is constant and W shows temperature dependence.

5. **Conclusion**
The observation of slip patterns produced by Vickers diamond indenter on cube-textured Al foils showed the curious effect that for indents with the diagonal parallel to cube axis deformed with a more or less square impression whereas that for one rotated at 45° showed a concave shape with spearhead shape at
the corners. The temperature dependence of the pattern showed that hardening mechanisms were
temperature dependent but did not change for the 45° orientation. However for the RD one, the near-
isotropic deformation behaviour at 80°C is taken to mean that SFT are no longer formed and are replaced
by Stroh-Hirth locks. The lattice rotation due to the formation of indents can be detected by SAXS as it
leads to streaking whenever the DBS condition is satisfied.

![Figure 5. Schematic diagram of SAXS set-up with rotatable specimen holder. Left image shows 2-D SAXS image at 0° position and one with streaks at the 12° position.](image)

References

[1] E. Demir, D. Raabe, N. Zaafarani, S. Zaefferer, Acta Mater. 57 (2009) 559-569.
[2] C.C. Tasan, M. Diehl, D.Yan, C. Zambaldi, P. Shamthraj, F. Roters, D. Raabe, Acta Mater. 81 (2014)
386-400.
[3] L.M. Brown, Mater. Sci. Forum, 550 (2007) 105-117
[4] K.K. McLaughlin, N.A. Stelmashenko, S.J. Lloyd, L.J. Vanderperre, W.J. Clegg, Mater. Res. Soc. Symp. Proc. 842 (2005) R1.3.1-R1.3.6.
[5] M.A. Singh, S. Saimoto, M.R. Langille, J. Lévesque, K. Inal, A.R. Woll, Philos. Mag. 97 (2017)
2496-2513.
[6] S. Saimoto, Low temperature deformation of copper single crystal oriented for multiple slip, Ph.D.
thesis, Massachusetts Institute of Technology, Cambridge, MA. USA, (1964).
[7] B.H. Kear, Trans. Metall. Soc. AIME, 224 (1962) 674-677.
[8] L.E. Collins, Plane strain deformation and subsequent recovery of ideally oriented copper single
 crystals, M.Sc. Thesis, Queen’s University, Kingston, Canada (1977).
[9] S. Saimoto, Philos. Mag. 86 (2006) 4213-4233.
[10] A. Akef, J.H. Driver, Mater.Sci. Engng. A 132 (1991) 245-255.
[11] C. Maurice, J. H. Driver, Acta metall. mater. 41 (1993) 1653-1664.
[12] C. Xu, Y. Zhang, F. Lin, G. Wu, Q. Liu, D. Juul-Jensen, Metall. Mater. Trans. 47A (2016) 5863-
5870.
[13] Y. Nakata, Philos, Mag. 11 (1965) 251-261.
[14] G. Langelaan, S. Saimoto, Rev. Sci. Instrum. 70 (1999) 3413-3417.
[15] R. Le Hazif, J-P. Poirier, Acta Metall. 23 (1975) 865-871.
[16] H. Ohkubo, Y. Shimomura, I. Mukouda, K. Sugio, M. Kiritani, Mater. Sci. Eng. 350 (2003) 30-36.

Acknowledgements

The authors thank Cornell High Energy Synchrotron Source (CHESS) at which the SAXS studies were
performed and the Natural Sciences and Engineering Council of Canada for support over many years.