Microstructural self-organization triggered by twin boundaries during dry sliding wear

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Abstract

A novel microstructural self-organization reaction is observed near the surface of a Cu–Ni–Sn bronze subjected to dry sliding wear. Scanning electron microscopy using electron-backscattered diffraction reveals the formation of near periodic patterns comprising domains with alternating orientation separated by narrow boundaries. Remarkably, orientation patterning takes place only near twin boundaries intercepting the sliding surface, and only on a specific side of the boundaries. The domain period, which ranges from a few to tens of micrometers, increases with the applied load. This orientation patterning is suppressed by promoting steady sliding using solid lubricants such as Ag and self-generated oxide-bearing tribolayers. Transmission electron microscopy reveals planar dislocation glide in the orientation domains, with multiple active slip systems. A rationalization of these results is offered based on an instability triggered by the interaction of elastic stick–slip waves with pre-existing twin boundaries.

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1. Introduction

Plastic deformation drives crystalline materials into nonequilibrium states, often leading to the formation of self-organized microstructures [1–3]. These structures are observed during testing under monotonic and cyclic loading [3], and in service, e.g. during sliding wear [4–6]. A key factor responsible for the stabilization of these self-organized structures is the interaction between dislocations through their elastic fields. Owing to these interactions, a homogeneous dislocation density may be unstable and evolve into patterns comprising domains with low dislocation density, e.g. cells or cell blocks, separated by regions of high dislocation density, e.g. cell walls and geometrically necessary boundaries (GNBs) [7–9]. In the later case, GNBs separate lamellar cell blocks with alternating crystallographic misorientations [10,11]. The GNBs are usually parallel to the main direction of plastic flow, as observed in rolling [10] or during frictional sliding [5]. Furthermore, the average domain size and misorientation across GNBs increase with strain, and the distributions of domain size and misorientation obey similitude relationships as first reported by Hughes [12]. It has been proposed that the formation of lamellar cell blocks results from the difference in activity of slip systems in the regions abutting a GNB [13–15]. In addition to a fundamental interest in understanding the conditions leading to self-organized structures, a practical motivation is that the unique microstructures stabilized by self-organization reactions often result in modifications of the properties of these materials, in particular their mechanical properties. These modifications can either be detrimental or beneficial to the performances of these materials. A better understanding and control of self-organization reactions may thus provide a path for improving materials performances.

An outstanding question regarding self-organization induced by plastic deformation is to evaluate the role that
grain boundaries play on the stabilization of domain structures. As established by Livingston and Chalmers [16], the plastic strain incompatibility at grain boundaries can be accommodated by activating additional slip systems in the regions adjacent to the grain boundary (GB). It is therefore not surprising to observe microstructural heterogeneities such as orientation gradient and lattice curvature in the vicinity of GBs in polycrystalline materials subjected to plastic deformation [17–20]. It remains difficult, however, to anticipate the impact of specific GBs on overall deformation structures and local grain orientations. For instance, Zaefferer et al. [21] performed channel die compression tests of three aluminum bicrystals with different symmetric \( \langle 112 \rangle \) tilt boundaries and found that the deformation behavior of the bicrystals resembles that of the two individual single crystals. They also showed that a crystal plasticity finite element model predicts intragranular strain heterogeneity that compares very well with the experimental data, even though the model does not include any grain boundary contribution. Similarly, Xuang and Winther [9] reported that in Al and Cu deformed in tension or by cold rolling, grain boundaries, including annealing twin boundaries, did not perturb the morphology of GNBs. In another study, however, Dorner et al. [22] showed that in Fe–3 at.% Si steel deformed in compression to a strain of 36%, grain boundaries triggered the formation of orientation domains, 2–3 \( \mu \)m in size, which formed within microbands that initiated at GBs. The misorientations between adjacent domains were large, up to 25\(^\circ\). Moreover, these domains were organized into quasi-periodic patterns, with periodicity \( \sim 2–4 \mu \)m. A rationalization for the formation of orientation domains was offered based on local variations in active slip systems due to work hardening, although the specific mechanisms leading to the stabilization of quasi-periodic domains and selecting the characteristic patterning length remained elusive.

In the present work, we report on a related but distinct microstructural self-organization in materials subjected to dry sliding wear. This novel reaction leads to the spontaneous formation of near-periodic structures comprising orientation domains separated by well-defined boundaries. Quite remarkably, the domain patterns, observed here in a Cu–Ni–Sn bronze, form only on one specific side of pre-existing twin boundaries, and only for wear test conditions favoring stick–slip sliding. A rationalization is proposed based on the interaction between stick–slip waves and pre-existing twin boundaries.

2. Experimental procedure

A spinodally hardened Cu–15 wt.% Ni–8 wt.% Sn alloy (hereafter referred as CuNiSn) was subjected to sliding wear using a Koehler K93500 pin-on-disc tester. Cylindrical pins, 6 mm in diameter and 60 mm in length, and 6 inch diameter discs were machined by electrical discharge machining (EDM). Dry sliding wear tests are performed at room temperature (RT), in air, under 0.1–10 kgf load, 0.05–0.75 m s\(^{-1}\), and for 2–4 h to reach steady state wear. Prior to testing, the contacting surfaces of the disc and pins were polished using SiC grinding papers with grit sizes down to 1200 grit to achieve an average surface roughness \( (R_s) \) less than 1 \( \mu \)m. The wear rates have been estimated by direct measurement of the weight loss of the pin to an accuracy of \( \pm 0.1 \) mg as well as from the linear displacement vs. time curves recorded during the wear test by a linear variable displacement transducer (LVDT). Unless stated otherwise, the same bronze material was used for the disc.

For characterizing grain orientation, a laboratory frame of reference was defined by the sliding direction (SD), the transverse direction (TD) in the sliding plane, and the direction normal to the wear surface (ND). After wear testing, the pins were cut for cross-sectional scanning electron microscopy (SEM) observation in the SD–ND plane. The worn surface of the pin was coated with 20–30 nm of Ni to protect the worn surface during polishing. Microstructural characterization and orientation mapping were performed in a JEOL 7000F SEM equipped with a field emission gun and an HKL Technology electron backscatter diffraction (EBSD) system. Orientation maps were acquired using a voltage of 25 kV, and a step size of 10 to 50 nm. Transmission electron microscopy (TEM) samples were prepared from the SEM cross-sections by the standard lift-out technique using focused ion beam (FIB), and these SD–TD samples were analyzed using a JEOL 2010F TEM operated at 200 kV.

3. Results

3.1. Microstructure of as-received CuNiSn alloy

The as-received CuNiSn alloy is a single-phase face-centered-cubic (fcc) material. The EBSD map in Fig. 1 shows
that it consists of nearly equiaxed grains with a mean grain size $\sim 150 \mu m$, a large fraction of annealing twin boundaries, and no preferred crystallographic orientation. The twin boundaries (TBs) were identified using EBSD as boundaries separating grains with a $60^\circ \pm 5^\circ$ misorientation around a $\langle 111 \rangle$ direction. The average twin thickness was 12 $\mu m$, covering an area fraction of $\sim 26\%$, with TB plane traces randomly oriented.

X-ray diffraction confirms the presence of a single fcc phase with spinodal decomposition in the as-received alloy while TEM diffraction patterns indicate the occasional occurrence of an ordered D$_{022}$ phase. For Cu–15 Ni–8 Sn, it has been established that at temperatures below 520 $^\circ C$, the alloy first undergoes spinodal decomposition leading to composition modulations with alternating Sn-rich and Sn-lean regions at a scale of several nm [23]. With increasing annealing time, the Sn-rich regions transform to the D$_{022}$ ($Cu_{Ni_{1.3}}$)$_3Sn$ ordered phase (for details, see the TTT diagram in Fig. 17 in Ref. [23]). The occasional D$_{022}$ precipitates present in our samples could not be resolved by the dark-field (DF) images, due to their small size and the strain contrast produced by the spinodal decomposition. Moreover, these precipitates represent a small volume fraction and are not found in all regions analyzed by TEM. These precipitates are thus not considered in the presentation and discussion of our results.

3.2. Tribological behavior of CuNiSn alloy

Fig. 2 reports the steady state wear rate and friction coefficient of the CuNiSn alloy tested under 0.1–10 kgf normal load, which translates into $\approx 0.35–3.5$ MPa nominal contact pressure, and 0.05–0.75 m s$^{-1}$ sliding velocity. The wear rates are of the order of $10^{-6}–10^{-5}$ mm$^2$, and friction coefficients are $\sim 0.5–1.0$ except for the lowest load. Both the wear rate and the friction coefficient are typical values for metals subjected to dry sliding wear [24]. Archard’s law [25] appears to hold approximately in this load range as the wear rate increases nearly linearly with applied load, although a power-law fitting yields a coefficient of 0.75, instead of unity. The friction coefficient reaches a high value of 3.2 at the lowest applied load of 0.1 kgf, which can be rationalized by the strong elastic deformation at contacting asperities at this low load [26].

3.3. Strain profile measurements

Plastic deformation near the worn surface was evidenced and measured by the displacement of grain boundaries. Pre-existing grain boundaries with long straight traces in the cross-sections observed by SEM were used as markers to calculate the strain profile during wear tests, following the standard marker displacement technique [27,28]. Fig. 3a shows an example of grain boundary deflection along the SD of a worn sample tested under 0.5 kgf normal load. An excellent fit of the grain boundary displacement (indicated by the arrow in Fig. 3a) vs. depth is obtained using an exponential decay, in agreement with prior studies [29]. The equivalent von Mises strain at a depth $x$ is then calculated as $e_{VM} = (dy/dx)/\sqrt{3}$, where $dy/dx$ is the slope of the displacement ($y$) vs. depth ($x$) curve. Fig. 3b shows the strain profiles measured using both general grain boundaries and twin boundaries. It can be seen that the strain profiles obtained from these different grain boundaries are nearly identical. The maximum equivalent strain, which is reached at the sliding surface, is measured to be $\sim 180\%$, or 310% shear strain since the equivalent strain $e_{VM}$ is related to the shear strain $\gamma$ by $e_{VM} = \gamma/\sqrt{3}$. The fact that the maximum strain is reached at the sliding surface is consistent with stress state calculated from contact mechanics for sliding contacts [30]. These calculations, using Tresca’s criterion for plastic yielding, indicate that the shear stress is maximum at the sliding surface when the coefficient of friction exceeds 0.3, which is the case for all wear tests reported here.

Table 1 lists the maximum strain and decay length for worn samples tested under applied load up to 10 kgf. The decay length of the exponential increases from 4.5 $\mu m$ for a load of 0.5 kgf to 28.6 $\mu m$ for a load of 10 kgf. The maximum equivalent strain, obtained by extrapolating the exponential fit of the strain-depth profile up to the sliding surface, is found to be $\sim 1.8–2.1$, nearly independent of the applied load. Using the measured steady-state wear rate, the strain-rate profile can be directly obtained from a given strain profile [31]. The maximum strain rates are found to be $\sim 10^{-3}$ to $5 \times 10^{-3}$ s$^{-1}$ for all the samples studied here (see Table 1 for details).

3.4. Self-organized orientation domains near twin boundaries

We first focus on a sample that was worn under 0.5 kgf normal load and 0.05 m s$^{-1}$ sliding velocity. Away from grain boundaries, deformation along the SD was homogeneous at the resolution provided by EBSD, $\sim 10$ nm, as illustrated in Fig. 4a. Homogeneous deformation and absence of grain re-structuration were also observed in sin-
The orientation patterning extends over 10–30 μm along the wear surface, as illustrated in Fig. 6. The penetration of patterning below the wear surface decreases as one moves away from its associated twin boundary. This patterning consists of domains of alternating crystallographic orientations, separated by fairly sharp boundaries. A misorientation profile from A to B in Fig. 6b measured with respect to the original point A shows a quasi-periodic arrangement of subgrain boundaries with a wavelength of ~3 μm. These boundaries separate domains that experienced rotation of smaller (see black arrowhead in Fig. 6a) and larger (red
arrowhead in Fig. 6a) amplitudes. The misorientation across these boundaries ranges from a few degrees to 35°.

Sliding wear imposes a simple shear deformation along the SD, which is expected to produce a rigid body rotation around the TD. This rigid rotation alone would result in crystallographic poles coming down along the SD in SD–TD pole figures. Fig. 6a shows that grain rotation in the “red arrow” domain is around the TD, thus largely
following the imposed rigid body rotation, while that in the “black arrow” domain is mainly around the ND. This indicates that the plastic spin generated by plastic deformation in that region was large enough to compensate for the rigid body rotation.

Quite remarkably, in the vicinity of some twin boundaries, grains were even observed to rotate against the rigid body rotation, as illustrated in Fig. 7. Fig. 7a shows an EBSD map at a twin boundary with orientation patterning in the top grain. Along line A, the grain rotation shows a discontinuity in the pole figure (see blue arrows in Fig. 7c) due to the formation of a low angle grain boundary at ~4.5 μm below the worn surface, see blue arrow in Fig. 7b. Along line B, the local orientation first follows the rigid body rotation but then it changes abruptly to the opposite direction (see red arrow in Fig. 7c), again at ~4.5 μm below the worn surface. At that depth, the grain boundary misorientation changes from 60° to ~50°, as indicated in Fig. 7b. The misorientation, however, goes back to 55–60° at ~3 μm below the worn surface. These observations suggest that, for this TB and the abutting grains, the boundary constrained the deformation of the grains it separates to the point that these grains rotated locally against the rigid body rotation so as to maintain a twin orientation relationship.

Fig. 8 shows similar microstructural self-organization in four other samples, tested under different loads. Orientation patterning is observed in all cases, independent of the orientation of the grains. The quasi-period of the orientation domain structure, measured from at least three different grains for each wear test condition, increased from ~3 μm at 0.1 kgf to 15 μm at 10 kgf, as shown in Fig. 9. Similar orientation patterning was also observed at TBs when the sliding velocity was varied from 0.05 to 0.75 m s⁻¹, but varying the sliding velocity did not affect the quasi-period of the patterning.

3.5. Sliding tests at elevated temperatures

All the results reported so far were from wear tests performed at or near room temperature. The wear tester used in this study is equipped with a heating stage, regulated by a thermocouple inserted into the pin ~4 mm away from the sliding surface. Steady-state temperatures measured in the previous tests never exceeded 50 °C. Additional experiments were then conducted by heating the pin to 80 °C and to 330 °C, using our reference (i.e. self-mating) conditions, for a few selected values of load (0.5 and 10 kgf) and sliding velocities (0.05 and 0.75 m s⁻¹). Microstructural self-organization was observed for all tests run at 80 °C, but never for the tests performed at 330 °C. At this elevated temperature, the microstructure just below the sliding surface instead comprises multiple grains with size varying from tens of nanometers to a few micrometers, and possessing a wide range of crystallographic orientations, see Fig. 10. This microstructure is characteristic of a material that has undergone recrystallization.

3.6. Sliding tests with solid lubrication

It is commonly observed during solid friction that steady sliding may be unstable [32]. Specifically, stability analysis of elastic solids in contact with moving obstacles shows that, for a large domain in the load and sliding velocity parameter space, steady sliding is unstable and replaced by stick–slip sliding [33–36]. The large coefficient of friction measured in our experiments, ranging from 0.5 to 1.0, as well as the large amplitude of the fluctuations of the coefficient of friction, indicate that, during the tests reported so far, stick–slip sliding was taking place. For instance, for a load of 0.5 kgf and a sliding velocity of 0.25 m s⁻¹, the variance of the friction coefficient fluctuations reaches the high value of 0.051, see Fig. 11a. In order to suppress stick–slip sliding and to investigate its possible
role on orientation patterning, additional experiments were carried out using solid lubricants. Two approaches were used to that end. In the first approach we changed the counterface material from a bronze disc to a martensitic stainless steel (SS440C), since previous studies [37] showed that in that case a lubricating metal-oxide nanocomposite tribolayer forms at the contacting surfaces. In the second approach, we kept the bronze disc as counterface but employed a split-pin geometry [38], and sandwiched a ~250 μm Ag foil between two halves of bronze pins. Ag was chosen since it is commonly used as a solid lubricant for sliding contacts from room temperature to moderately elevated temperatures. As shown in Fig. 11a, both approaches led to a reduction of the average coefficient of friction by a factor of 2. More importantly, the standard deviation of the friction coefficient fluctuations in the steady-state regime was reduced from 0.051 for the reference test conditions to 0.017 for the SS440C disc, and to 0.0085 for the split pin with Ag foil. These results are clear indications that both lubrication methods suppressed significantly unstable sliding. Therefore, while lubrication may not be a general solution to suppress stick–slip sliding [39], it did accomplish that for the present wear tests. Remarkably, in both cases, no orientation patterning was observed near twin boundaries. This is illustrated in Fig. 11b for the Ag lubrication method. We thus conclude that the orientation patterning does not take place when the sliding is steady. We also measured the strain profiles for these two modified wear tests and the maximum von Mises strain at the surface was 1.8 for the bronze pin running against SS440C, and 0.6 for the split pin with Ag foil, both determined under 0.5 kgf normal load. The strain profiles are similar to the ones measured for the self-mated tests, especially for the SS440C counterface, and thus the absence of patterning in the lubricated cases cannot just result from a change in the plastic strain near the wear surface.
that, in the absence of these solid lubricants, stick–slip sliding was taking place. Remarkably, when steady sliding was achieved using solid lubricants, orientation patterning did not take place. It can thus be concluded that the reported orientation patterning requires both pre-existing twin boundaries and stick–slip sliding. We now analyze and discuss these results in detail.

We first focus on the microstructures resulting from orientation patterning. The boundaries separating the self-organized domain structures are rather narrow, with fairly sharp misorientation gradients below the wear surface. Fig. 12 shows misorientation angle profiles measured as a function of depth from the sample tested under 0.5 kgf load and 0.05 m s\(^{-1}\) sliding velocity. The average misorientation increases from \(\sim 5^\circ\) at a depth of 6 \(\mu\)m to \(\sim 40^\circ\) right below the sliding surface, approximately following an exponential decay with a 2.7 \(\mu\)m decay length. Taking advantage of our measurements of strain vs. depth (Fig. 3b) and misorientation vs. depth (Fig. 12a), we can eliminate the depth variable and plot directly the misorientation vs. strain (Fig. 12b). As the domain boundaries are first observed at a finite depth below the wear surface, about 6 \(\mu\)m in Fig. 12a, the corresponding strain was subtracted when plotting misorientation vs. strain. As seen in Fig. 12b, a power law with exponent \(\sim 1\) fits these data quite well. Similar power law dependences have been reported for GNBs in fcc metals subjected to cold rolling [11,12,40] or to high pressure torsion testing [41], with power law exponents varying between 0.5 and 1. Analytical models of domain boundaries formed in materials subjected to severe plastic deformation (SPD) indicate that an exponent 1 is expected in the case when domain boundaries result from an imbalance of activity of slip systems between the regions abutting the boundaries [13,42]. The power law fit in Fig. 12b therefore suggests that this mechanism may also be responsible for the formation of the boundaries separating orientation domains in our observations. We emphasize that, while the present GNBs share similarities with the ones previously reported in the literature, they possess two very distinct characteristics. Firstly, the boundary plane traces tend to be parallel to the trace of the adjacent TB that triggered the patterning, whereas after rolling [43], friction test [5], and sliding wear [6,44], GNBs traces were found to be aligned with the main material flow direction. Secondly, the width of the orientation domains does not decrease noticeably with strain, in contrast to these prior reports.

In order to characterize the microstructure of orientation domains at a smaller scale, transmission electron microscopy samples were prepared from regions first selected by EBSD, using FIB and the lift-out technique. One such region is shown in Fig. 13a. It presents the advantage of containing a nanotwin in the patterning area, between the boundaries labeled TB5 and TB6 in Fig. 13a. EBSD maps indicate indeed that, far from the wear surface, the crystallographic orientation inside this lamella matches the one inside the grain on the left of the boundary that triggered the patterning instability, labeled TB3 in Fig. 13a. In addition, the three boundaries (TB4, TB5,
TB6) were identified as twin boundaries based on the four degrees of freedom that can be accessed by 2-D EBSD. Thanks to the presence of the nanotwin, these twin boundaries as well as the microstructure of orientation patterning can be conveniently investigated in one TEM sample. The white dashed-line rectangular box in Fig. 13a represents the Pt line deposited prior to the lift-out process, and it thus gives an approximate location of the TEM thin foil. This particular sample was taken below the sliding surface, which corresponds to an equivalent strain of 0.6. We note that, because of the procedure followed here to prepare TEM samples, it is an SD–TD cross-section. This allows information to be accessed along the TD, information that was missing from the 2-D EBSD maps.

Fig. 13b is a montage of 19 bright-field TEM images covering nearly 20 μm along the SD. Several grain and domain boundaries are clearly identified, including the grain boundary TB6, separating the nanotwin from the matrix, and the domain boundaries DB1, DB2, DB3 and DB4. We first discuss the boundary TB6. The TEM montage reveals that, in the SD–ND plane, the trace of this boundary is almost aligned with the TD. Precisely, its normal is rotated by 3° counterclockwise from the SD axis. On the ND–SD plane, the twin boundary plane normal is oriented 69° counterclockwise from the ND axis. The direction of the TB6 habit plane as well as the local orientations taken at points A and B abutting TB6 (shown in Fig. 13b) are plotted in the {111} ND–SD pole figure in Fig. 14a. These two local orientations A and B exhibit a 58° misorientation around a common ½112 axis and the TB6 plane normal deviates from this common ½112 direction by 3°. This establishes that the TB6 boundary, 5 μm below the wear surface, is within a few degrees from a symmetric, coherent twin boundary orientation. Since successive twin lamellae are parallel, see Fig. 1, we conclude that the TB4 habit plane has a similar orientation, and therefore that TB4 is also very close to a coherent twin boundary orientation, as previously inferred based on 2-D EBSD results.

Turning now to the domain boundaries DB1 to DB4, it is observed that their trace in the SD–TD plane of the TEM sample is predominantly parallel to the trace of the TB6 boundary, with a maximum misorientation of 25° between DB3 and TB6. This result, combined with the EBSD results in the SD–ND plane, confirms that the domain boundaries produced by orientation patterning tend to align along the twin boundary that triggered self-organization. In agreement with EBSD orientation maps obtained from other areas (Figs. 5–8), Fig. 14b and c shows that the local orientation obtained from EBSD along the wear surface (points C through I) alternates back and forth, through rotations that are primarily around the TD. Inside the orientation domains, significant dislocation activity is observed in planar segments extending over several micrometers, see Fig. 13b. This dislocation arrangement and their TEM contrast are similar to those...
reported by Büscher et al. in worn austenitic nitrogen–manganese steels [45], and are indicative of high dislocation density. As cross-slip is suppressed in low stacking fault energy (SFE) materials, such as the CuNiSn bronze investigated here and manganese steels, planar glide of dislocations is indeed expected. In contrast, in materials with moderate or large SFE, cross-slip stabilizes three-dimensional dislocation structures. For pure Cu, for instance, Heilmann et al. [6] reported the formation below the worn surface of cellular dislocation structures, elongated in the SD and only a few hundreds of nanometers wide, and similar structures have been found by Walker et al. in aluminum alloys [46]. Thanks to planar dislocation glide in the CuNiSn bronze, a lower bound for the number of active slip systems is easily obtained. Fig. 15a is a bright field image taken from point J, after rotating the sample by $19^\circ$ so as to bring it to the $110$ zone axis. Slip traces are observed in all three $\{110\}$ directions normal to $111$, establishing that at least three slip systems are locally active. For one of these slip systems, the Schmid factor is very low if we only consider the applied shear stress along the SD, see Table 2. The activation of this system therefore requires the presence of large internal stresses. Note that slip activity in the $\{111\}$ plane, which is nearly parallel to the sliding surface, cannot be easily detected here since the TEM sample is an SD–TD section. Slip activity in that plane is, however, likely to take place since it contains slip systems with high Schmid factors. It is also observed that slip traces rotate when they cross domain boundaries, as illustrated in Fig. 15b at the DB4 boundary. From EBSD data, the misorientation between points H and I is calculated to be $18^\circ$, but the misorientation between the $\{110\}$ directions projected on the SD–TD plane is $8^\circ$. This is in good agreement with the rotation of $6^\circ$ of the $\{110\}$ slip trace measured from the TEM image. The cutting of domain boundaries by slip traces generates steps that can reach 50–100 nm, see Fig. 15b, thus requiring the glide of hundreds of dislocations in these planes. Finally we note that it is difficult to image single dislocations owing to the strain contrast produced by wear and by spinodal decomposition (see for instance Fig. 1 in Ref. [37]). It is thus not possible to analyze dislocation nature and content in more detail.

We now discuss the origin for the asymmetry of the patterning, that is, the fact that it is only observed on the leading side of the triggering TB. We propose that this asymmetry results from the interaction of pre-existing TBs with stick–slip waves generated by unsteady sliding. Indeed, during unsteady sliding, elastic stick–slip waves propagate at the interface between the two contacting bodies, similar to Schallamach waves [32,39,47]. In the case of a pin sliding against a flat, which corresponds to the geometry of our wear testing, an important characteristics of stick–slip separation waves is that they initiate at the trailing end of the pin and propagate toward its leading end [48,49]. This is due to the fact that tensile stresses are

Fig. 14. (a) $\{111\}$ ND–SD pole figure of orientations of A and B, and plane normals of TB6, DB1–4 as labeled in Fig. 13b. (b) $\{110\}$ ND–SD pole figure of orientations of C to I, as labeled in Fig. 13. (c) Enlarged box area in (b).

Fig. 15. Bright field TEM images of (a) box J and (b) box K, as shown in Fig. 13b. The scale bar in both images is 1 $\mu$m.
first generated at the back end of the slider. Therefore, when considering two grains separated by a twin boundary intercepting the sliding surface, the plastic deformation of the trailing edge grain should not be affected by the presence of the TB since stick–slip waves encounter the boundary after traveling through that whole grain. In contrast, stick–slip waves interact with that TB before propagating into the leading edge grain and we propose that this interaction is responsible for the formation of patterns of orientation domains. It is interesting to notice that static restoration mechanisms near $\Sigma 3$ boundaries have been observed in plastically deformed materials. For instance, Thomson and Randle reported that in Ni, while straining reduced the fraction of $\Sigma 3$ boundaries with exact coincidence site lattice (CSL) matching, intermediate temperature annealing led to an increase of this fraction [50]. SEM-EBSD results (Fig. 7) and TEM results (Fig. 13) show that the regions adjacent to TBs tend to deform in a way that maintains, approximately, a twin orientation relationship. This suggests that a dynamic restoration mechanism is active. Maintaining a twin orientation is, however, only possible locally since the lattices of the two grains abutting the boundary rotate differently, owing to the difference in activity of their slip systems. In particular, the leading grain near the twin boundary would experience a lattice rotation incompatible with the bulk rotation for that grain. In order to compensate for this incompatibility, a second region with a distinct orientation and a distinct lattice rotation rate must form, as we observed experimentally, see Figs. 5–8 and 14b and c. The formation of alternating orientation domains is in fact commonly observed across GNBs [10], and kinematics modeling [15] indicates that such a microstructural organization is an effective way for crystals to accommodate large plastic strain.

The above analysis, while providing a rationalization for the asymmetric formation of alternating orientation domains in the vicinity of twin boundaries, does not explain directly the stabilization of quasi-periodic patterns. However, recent modeling efforts on dynamic instabilities in crystals driven by plastic deformation point to the stabilization of periodic structures. In particular, Prantil et al. [51] studied the dynamic nature of preferred orientations in planar crystals. They found that grain rotation can be either monotonic or periodic, depending on the relative magnitude of the stretching and spin rates, and slip system geometry. Tonks et al. [52] pointed out that these planar crystals rotate in a manner similar to coupled oscillators in Kuramoto’s model. Using crystal plasticity finite element modeling they identified a domain in parameter space where the crystal develops near periodic patterns of orientation domains. Similarly to our experimental observations, the domains are separated by fairly narrow boundaries. In the work of Tonks et al. [52], patterns were observed to form spontaneously in an initially defect-free material. This is different from our work since, away from twin boundaries, we did not observe pattern formation. This difference is, however, not prohibitive since in dynamical systems that are stable but close to the onset of an instability, the introduction of a defect, a TB in our case, can trigger this instability [2]. The work of Gavrielides et al. [53] on lattices of coupled chaotic oscillators illustrates this point: in the presence of site impurity, the dynamical behavior of the oscillators can lock into periodic spatiotemporal patterns, even though the forcing conditions were such that a spatially homogeneous response was observed in the absence of impurity. Lastly we note that our results point to the importance of intermittency of plastic deformation on microstructure evolution. The cyclic nature of the deformation imposed by stick–slip sliding was shown in the present study to be a necessary condition for triggering orientation patterning. This suggests that orientation patterning may also develop during fatigue testing.

Some questions remain open and will require further investigation. Firstly, as for the instability reported by Dorner et al. [22], the processes responsible for selecting the period of the self-organized microstructure remain to be elucidated, although our measured values of 2–15 μm are similar to those reported for orientation domains in SPD materials [7,10]. Secondly, it will be interesting to fully identify the materials parameters required for microstructural self-organization to take place during sliding wear. The present results indicate already that, in addition to the presence of TBs and stick–slip sliding, the development of orientation patterning requires large initial grains and a material that resists dynamic recrystallization at the worn surface. For instance, wear tests performed with annealed pure Cu pins resulted in recrystallized surface microstructures, and not surprisingly, no periodic patterns of orientation domains were observed in that case.

### 5. Conclusion

Subsurface microstructure evolution in a Cu–15 wt.% Ni–8 wt.% Sn bronze after dry sliding wear test was studied by SEM-EBSD and TEM. Strain profiles were measured

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**Table 2**

Calculated Schmid factor of 12 $\{111\}/\{110\}$ slip systems based on local grain orientation in box J as shown in Fig. 13b.

<table>
<thead>
<tr>
<th>Slip system</th>
<th>Schmid factor</th>
</tr>
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<tbody>
<tr>
<td>(1–11)[011]</td>
<td>−0.0823</td>
</tr>
<tr>
<td>(1–11)[10–1]</td>
<td>0.8196</td>
</tr>
<tr>
<td>(1–11)[−1–10]</td>
<td>−0.7373</td>
</tr>
<tr>
<td>(111)[0–11]</td>
<td>0.0122</td>
</tr>
<tr>
<td>(1–11)[1–10]</td>
<td>0.0187</td>
</tr>
<tr>
<td>(111)[110]</td>
<td>−0.0309</td>
</tr>
<tr>
<td>(1–11)[011]</td>
<td>−0.0501</td>
</tr>
<tr>
<td>(1–11)[−10–1]</td>
<td>−0.2713</td>
</tr>
<tr>
<td>(1–11)[1–10]</td>
<td>0.3214</td>
</tr>
<tr>
<td>(111)[0–11]</td>
<td>−0.1019</td>
</tr>
<tr>
<td>(111)[10–1]</td>
<td>0.2862</td>
</tr>
<tr>
<td>(111)[−110]</td>
<td>−0.1843</td>
</tr>
</tbody>
</table>
using grain boundaries as markers. Strain increased exponentially toward the wear surface and reached a maximum of \( \sim 2 \), regardless of the applied load. For self-mated wear tests performed at room temperature, a novel quasi-periodic microstructural self-organization was systematically observed at pre-existing twin boundaries, but only in the grain on the leading edge side of the twin boundaries. The resulting orientation domains were separated by sharp domain boundaries, with misorientations ranging from 5° to 40°. Orientation patterning was no longer present when solid lubricants were used to achieve steady sliding, or when the wear tests were run at elevated temperature (330 °C), in which case recrystallization dominated the wear surface microstructure. This ensemble of results is rationalized by proposing that orientation patterning results from the interaction of pre-existing TBs with stick–slip waves generated during unsteady sliding. This rationalization explains in particular the asymmetry of orientation domain formation between the two grains separated by a TB. These results also point to the role that twin boundaries and intermittent deformation can play in the evolution of the microstructure of materials subjected to large plastic deformation.

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