

# Microstructure evolution in deformed copper

P. Landau · R. Z. Shneck · G. Makov · A. Venkert

Received: 30 April 2007 / Accepted: 17 July 2007 / Published online: 20 August 2007  
© Springer Science+Business Media, LLC 2007

**Abstract** The effects of strain and temperature on the microstructure and the detailed structure of dislocation walls in compressed pure polycrystalline copper were systematically studied. The microstructure observed consisted of dislocation cells and second generation microbands (MB2) and is presented in a microstructural map. The detailed structure of the cell's boundaries evolves with increasing strain and/or temperature from tangled dislocations into arrays of parallel dislocations. Interplay between strain and temperature controls the microstructure and the detailed structure of the dislocation boundaries. Above  $0.5 T_m$  only MB2 are observed; a different type of MB2 is also observed, one order of magnitude wider than the one observed at the lower temperature. With increasing strain and temperature the MB2 density increases. It seems that dislocation cells and MB2 are competitive dislocation patterns and at elevated temperatures the MB2 is the preferred dislocation pattern.

## Introduction

Patterning of dislocations into boundaries, walls and cells is common to many metals following plastic deformation. In particular, in fcc metals with medium or high stacking fault energies, a distinct cellular structure is formed at low

levels of strain. Several models have been proposed to explain the dislocation accumulation into dislocation boundaries and one of the most common is the low energy dislocation structure (LEDS) theory [1, 2].

The extensive research devoted to this phenomenon has revealed many of its characteristics [1–19]. In this paper we focus on the phenomenology of the microstructure evolution and detailed structure of dislocation boundaries in copper.

The microstructure observed following plastic deformation is highly dependent on the deformation mode [7, 8], strain rate [19] and temperature [6, 8]. Metallurgical factors such as grain size [3, 7], grain orientation [14, 15] and stacking fault energy [6, 8, 19] of the metal will also affect the microstructure. Furthermore, even the orientation of the section observed (longitudinal section, cross section) will reveal different dislocation patterns [14, 15].

In aluminum a small amount of strain is required for dislocations to rearrange into dislocation boundaries (DBs) delineating cell blocks (CBs) [1–10]. The DBs form misorientations between adjacent cells that increase with increasing strain. Several types of boundaries are distinguished including incidental DBs (IDBs), dense dislocation walls (DDWs) and geometrically necessary boundaries (GNBs) [1–5, 8, 9]. Further increase in strain will cause splitting of DDWs to form first generation microbands (MB1) that contain pancake shaped dislocation cells within them.

The microstructure of deformed copper is found to be more complex. The deformed grains were divided into two populations [16–18]: 1. Grains with large number of intersecting DDWs and MB1 ( $\sim 1/3$  of the grains) termed high wall density structures (HWD). 2. Grains with few DDWs and equiaxed dislocation cells ( $\sim 2/3$  of the grains) termed low wall density (LWD). Microbands of a different

---

P. Landau (✉) · R. Z. Shneck  
Department of Materials Engineering, Ben-Gurion University,  
P.O. Box 653, Beer-Sheva 84105, Israel  
e-mail: landaup@bgu.ac.il

G. Makov · A. Venkert  
Department of Physics, NRCN, P.O. Box 9001, Beer-Sheva  
84190, Israel

type, termed second generation microbands (MB2), were observed in the deformed grains. The grains with LWD structures have a high density of MB2 within them, compared to the HWD structures. However, in most reports regarding the microstructural features there is no distinction between the different types of grain populations [7, 9, 13–15, 19–22].

The MB2 are believed to have concentrated shear associated with them [15–26]. These MB2 were observed in nickel only following torsion [2] and were not formed in aluminum. The MB2 consist of double dislocation walls approximately 0.1–0.4  $\mu\text{m}$  apart and are formed on {111} planes above  $\sim 10\%$  deformation. They create small non accumulative misorientation ( $1\text{--}3^\circ$ ) [16–19]. Their density increases with increasing strain and they tend to cluster into groups, as wide as 8 nm [20, 22].

Several classifications have been introduced in the literature concerning the microstructure and dislocation patterns formed during plastic deformation [2, 7, 13–18].

The observed microstructures following tension in aluminum and copper were divided by Huang et al. [14, 15] into three groups: type 1: grains that are subdivided by long, straight DDWs on {111} planes; type 2: grains that are subdivided by ordinary DBs and form equiaxed cells in the cross section (perpendicular to the deformation direction); type 3: grains that are subdivided as for type 1, though, the DBs are not on crystallographic {111} planes. However, in this work the role of the MB2 was not discussed.

In many studies no distinction was made between MB1 and MB2 (for example: 23, 27–29), perhaps since it is difficult to distinguish between the two at high strain levels [2]. The role and importance of the MB2 is not clear, and whether they play an important role in the deformation has not yet been determined [2].

The detailed structure of dislocation walls was discussed in a few reports. The dislocation boundaries formed in aluminum during plastic deformation were found to consist of networks of parallel dislocations (several observations at room temperature [30, 31], mostly reported following creep and annealing [31]). The detailed structure of DBs in copper following room temperature deformation consists of tangled dislocations. Following deformation by creep the dislocations form networks though, not as regular as were observed in crept aluminum [32–35].

The differences between the deformed microstructures of aluminum and copper are often attributed to the lower stacking fault energy (SFE) of copper relative to aluminum ( $\sim 60$  and  $\sim 180$   $\text{mJm}^{-2}$  respectively) [11, 14, 15]. Lower SFE impedes cross slip and hinders the cell structure formation [6]. Another distinction between the two metals is the difference in the homologous temperature of the deformation. Increasing the deformation temperature for copper increases the mobility of dislocations.

In copper, the limited ability of dislocations to cross slip can be compensated by strain and the mobility of the dislocations can be enhanced by increasing the deformation temperature. Thus, enabling the formation of similar structures of dislocations such as those observed in aluminum.

The present work explores systematically the combined effect of strain and temperature on the microstructure and the detailed internal structure of dislocation walls in copper. We also compare the microstructures and detailed structure of DBs of copper to that of aluminum. The results are plotted onto a microstructural map manifesting the interplay between strain and temperature.

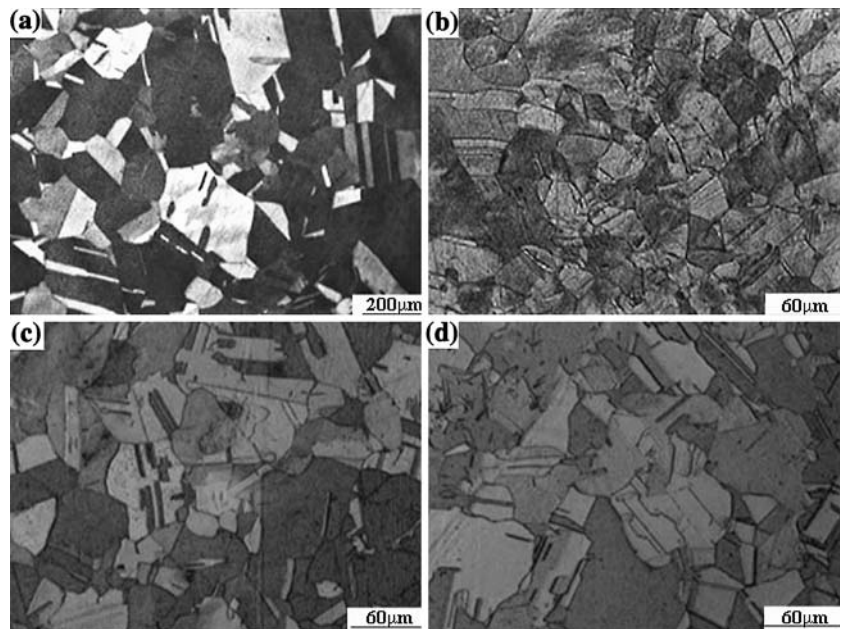
## Experimental

A high-purity polycrystalline copper (99.999%) was cut into cylindrical rods 1 cm long and 0.7 cm in diameter. The specimens were annealed at 500  $^\circ\text{C}$  for 5 h. The rods were uniaxially pressed in an Instron machine to different reductions (7, 10, 20 and 50%) at various temperatures (room temperature, 350, 500, 575 and 650  $^\circ\text{C}$ ). Metallographic and transmission electron microscopy (TEM) samples were sectioned perpendicular to the compression axis (similar to the transverse section of a rolled sample). The specimens were cut to  $\sim 200$   $\mu\text{m}$  slices by electro-erosion machine from which TEM samples, 3 mm diameter disks, were punched. Final thinning was achieved by jet electro-polishing. Ion milling (Gatan-PIPS<sup>®</sup>) was used to remove any contamination caused by the electro-polishing. The dislocation structures were examined in a Jeol-2000FX transmission electron microscope. Care was taken in order to avoid additional mechanical deformation during thinning. Examination of annealed reference samples confirmed that the sample preparation technique introduces little or no dislocations.

## Results

The combined effect of strain and temperature on the evolution of the microstructure and the different microstructural features was studied. From metallographic observations the initial grain size was found to be  $\sim 100$   $\mu\text{m}$ . Recrystallization was observed in all specimens following deformation at 500–575  $^\circ\text{C}$ , which produced a grain size of  $\sim 30$   $\mu\text{m}$ . At room temperature, dynamic recrystallization (DRX) was observed only following 50% deformation, as Belyakov et al. [36] reported, and the grain size was  $\sim 40$   $\mu\text{m}$  (Fig. 1). At higher temperatures (650  $^\circ\text{C}$ ) grain growth (grain size of  $\sim 70$   $\mu\text{m}$ ) and stepped annealing twins were observed. Deformation twins were

**Fig. 1** Metallographic micrographs showing the differences in grain size following compression in copper: **(a)** 10% strain at room temperature ( $\times 50$ ), grain size =  $98 \pm 27 \mu\text{m}$ . **(b)** 50% strain at room temperature ( $\times 200$ ) grain size =  $39 \pm 22 \mu\text{m}$ . **(c)** 10% strain at  $500^\circ\text{C}$  ( $\times 200$ ) grain size =  $30 \pm 14 \mu\text{m}$ . **(d)** 10% strain at  $575^\circ\text{C}$  ( $\times 200$ ) grain size =  $17 \pm 12 \mu\text{m}$



not observed. However, annealing twins were found following all strains at all deformation temperatures resulting from sample annealing prior to deformation.

The samples were sectioned perpendicular to the compression axis, which is similar to the transverse section of a rolled specimen, where the cells are seen equiaxed. However, in this section it was difficult to discern between DDWs, MB1 and GNBs.

TEM observations revealed that the microstructure evolves with increasing strain into a defined cell structure combined with MB2. There was only a slight (not significant) decrease in cell size with increasing strain and temperature. Above  $500^\circ\text{C}$  there is a transition to a dislocation pattern consisting solely of MB2. The evolution of the microstructure and the detailed structure of the different features are presented.

#### Room temperature deformation

At room temperature ( $0.22 T_m$ ), low levels of strain did not affect the grain size. However, following 50% strain a large decrease in grain size was observed, which we believe to be continuous DRX [36].

The microstructure is characterized by dislocation cells and MB2. The MB2 were observed above 10% strain as was reported previously [19]. With increasing strain the cells become more defined (sharper boundaries) and the density of MB2 increases, as was previously reported [22–25]. The MB2 width is  $0.1\text{--}0.5 \mu\text{m}$ .

Figure 2 illustrates the typical cell structure and MB2 observed by TEM in the copper specimens compressed to

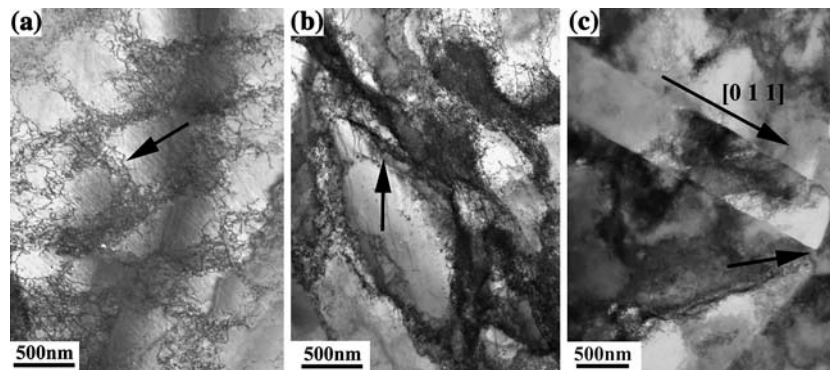
10, 20 and 50% strain. Following 10% strain a cell structure has already been formed and the DBs consist of tangled dislocations, which are arrowed in Fig. 2a. With increasing strain the cell boundaries are sharper (arrowed in Fig. 2b) and consist of parallel wavy dislocations. Cell size does not change significantly in this range of strains. This structure is consistent with the type 2 structure observed in copper following torsion and consists of ordinary equiaxed dislocation cells. The detailed structure of the cell walls consists of parallel dislocations. The dislocations form arrays with 2 or 3 different burgers vectors in each (for example Fig. 4a). Although the strain level is higher, there was no apparent decrease in the cell size, probably because the dislocations form recrystallized grain boundaries.

MB2 were observed above 7% strain. Their width remains  $\sim 0.2 \mu\text{m}$  at all strain levels and they consist of dislocations. Clustering of MB2 is observed with increasing strain. A MB2 intersecting a grain boundary, causing shear offset, following 50% strain is shown in Fig. 2c. The shear offset is arrowed.

#### Deformation at $350^\circ\text{C}$

Metallographic observations at  $350^\circ\text{C}$  ( $0.45 T_m$ ) showed that low levels of strain did not affect the grain size. Recrystallization was observed following 50% strain to yield a grain size of  $\sim 30 \mu\text{m}$ , similar to the observed grain size at room temperature.

Figure 3 illustrates the typical microstructure observed by TEM following deformation at  $350^\circ\text{C}$ . Following 10%



**Fig. 2** TEM bright field images showing the typical microstructure of copper observed following room temperature compression: (a) 10% strain—the cell walls (arrowed) consist of tangled dislocations. (b) 20% strain—the cell walls (arrowed) are sharper. No apparent

decrease in cell size is observed (c) 50% strain—A MB2 with [0 1 1] direction intersecting a grain boundary causing a shear offset, which is arrowed

strain a cellular structure combined with MB2 is observed. The MB2 are arrowed in Fig. 3a. The cell boundaries consist of tangled dislocations. Groups of MB2 are more frequent than at the same deformation level at room temperature. With increasing strain (20%) the dislocations forming the cell boundaries (arrowed in Fig. 3b) are wavy and parallel (detailed structure shown in Fig. 4b) and after 50% strain the dislocations form arrays of straight parallel dislocations (Fig. 4c). It can also be seen that the spacing between dislocations inside the DBs decreases with strain (comparing Fig 4b, c). The width of the MB2 does not change with increasing strain as can be seen in Fig. 3c. However, the density of MB2 increases with increasing strain, as well as with increasing temperature.

#### Deformation at 500 °C

Recrystallization was observed at all levels of strain at this temperature. The grain size was  $\sim 30 \mu\text{m}$ .

The microstructure formed during deformation at 500 °C ( $0.57 T_m$ ) consisted of dislocation cells combined with MB2. Two populations of MB2 were observed, distinguished by their widths. One of them ( $0.1\text{--}0.3 \mu\text{m}$  wide)

was also observed at the lower deformation temperatures; the other one order of magnitude wider ( $>1 \mu\text{m}$  wide). As strain increases, the density of both types of MB2 increases.

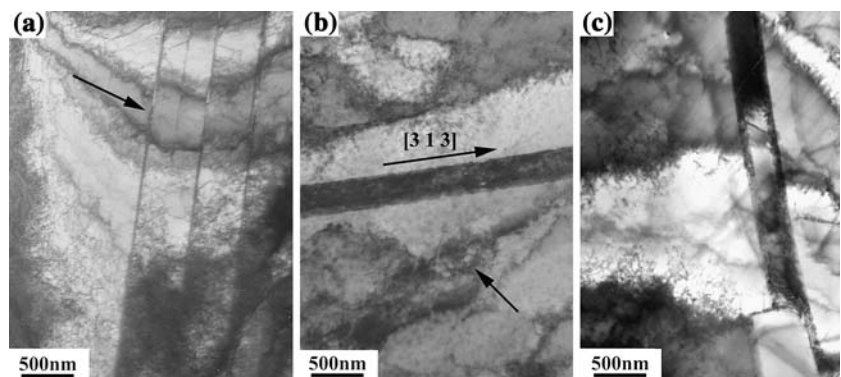
Figure 5 illustrates the typical microstructure observed following deformation at 500 °C. A cell structure was formed at all degrees of strain at this temperature (for example—cells arrowed in Fig. 5a). Clusters of MB2 were frequently observed. Following 50% strain the cell walls still consist of tangled dislocations. The density of MB2 increased with strain and temperature. Wider MB2 are observed in Fig. 5b.

#### Deformation at 575 °C and 650 °C

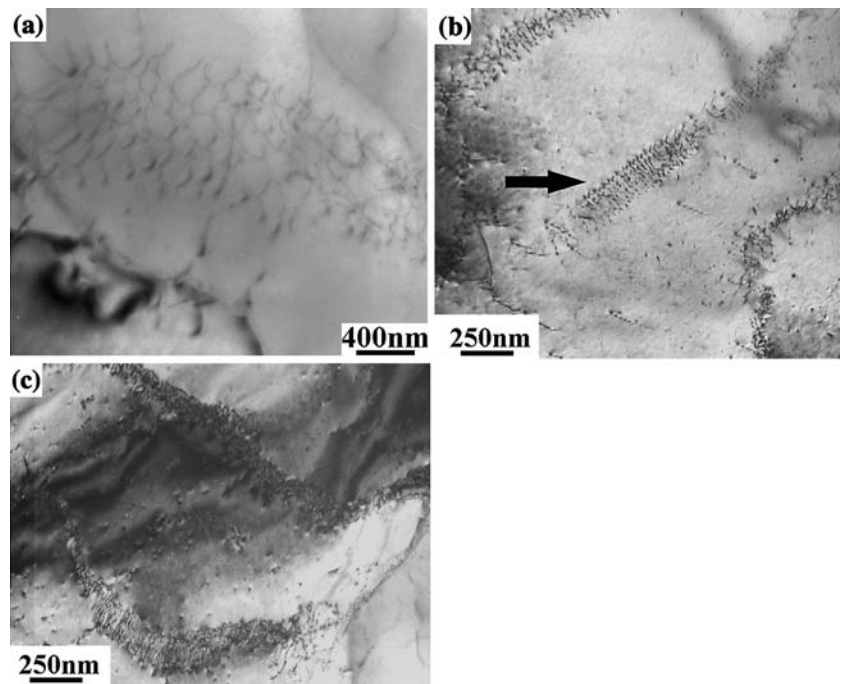
Metallographic observations revealed recrystallization at 575 °C at all degrees of strain, as was observed following deformation at 500 °C and the grain size was  $\sim 20 \mu\text{m}$ . At the higher temperature (650 °C) grain growth was observed, to a grain size of  $\sim 100 \mu\text{m}$ .

Following deformation at 575 °C ( $0.62 T_m$ ), at the lower strains, no dislocation cells were observed. Following 50% strain diffuse cells were observed. No dislocation cells were observed following deformation at 650 °C ( $0.68 T_m$ )

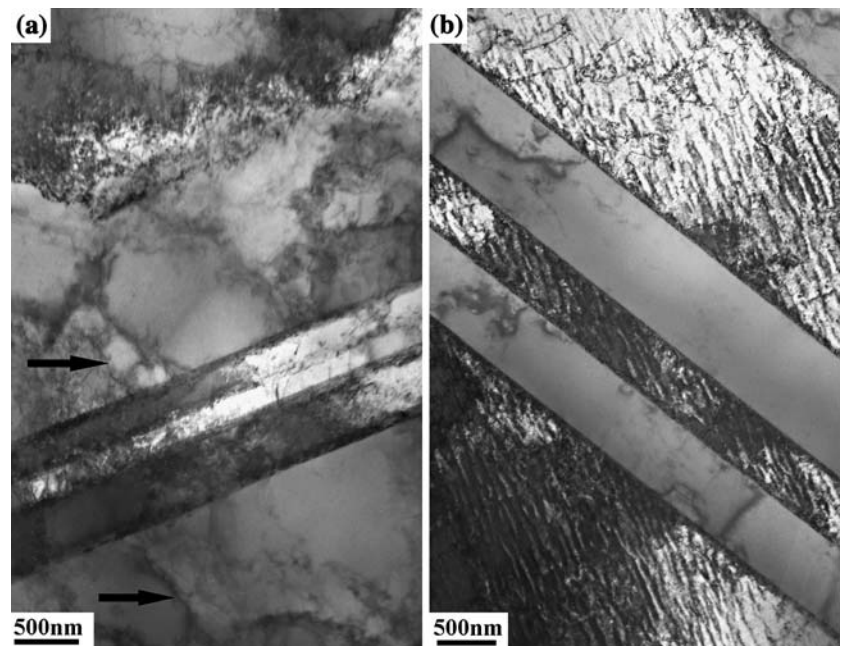
**Fig. 3** TEM bright field images showing the typical microstructural features observed following compression at 350 °C: (a) 10% strain—a cluster of at least 3 MB2 (arrowed). (b) 20% strain—A MB2 with the direction [3 1 3] is shown in the micrograph. Dislocation cell boundaries are arrowed. (c) 50% strain—showing MB2



**Fig. 4** Internal structure of DBs formed in copper following compression: (a) 50% strain at room temperature—the dislocations forming the DBs form ordered structures; a hexagonal network consisting of 3 different burgers vectors. (b) Internal structure of DB formed following 20% strain at 350 °C. The dislocations are parallel wavy (arrowed). (c) Internal structure of DBs formed following 50% strain at 350 °C. The dislocations form arrays of parallel dislocations



**Fig. 5** TEM bright field images showing the typical microstructure observed following compression at 500 °C: (a) 10% strain—clusters of MB2 are more frequent, combined with a cell structure. (b) 50% strain—wider MB2 are observed. Their width is one order of magnitude wider than those observed at the lower temperature deformation

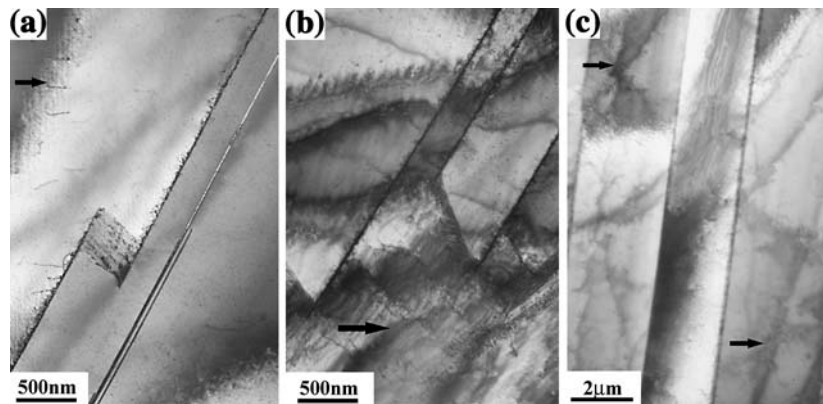


(Fig. 6). Following 50% strain the dislocations form tangles (arrowed in Fig. 6c).

The main microstructural features observed at elevated temperatures were the MB2, both narrow (0.1–0.3  $\mu\text{m}$ ) and wide (1–2  $\mu\text{m}$ ). With increasing strain the density of both types of MB2 increases. As strain increases the density of dislocations in the vicinity of the MB2 increases (Fig 6a and b).

## Discussion

The microstructure of deformed copper consists of dislocation cells and MB2 that evolve with increasing strain and temperature. Increasing strain gives opportunity for the dislocation to arrange into ordered structures, and increasing temperature raises the dislocation mobility. Both factors enhance the dominance of the MB2.



**Fig. 6** TEM bright field images showing the typical microstructure observed following compression at 650 °C. Only narrow and wide MB2 are observed. (a) 10% strain—cluster of 2 MB2. In the vicinity of the MB2 sparse dislocations are observed (arrowed). (b) 20%

strain—the density of thick and thin MB2 increased. The dislocations in the vicinity of the MB2 are wavy (arrowed) and their density increased. (c) 50% strain—the dislocations in the vicinity of the MB2 form tangles (arrowed)

A microstructural map is presented in Fig. 7, which summarizes the different dislocation patterns observed in copper and presents the evolution of the various dislocation patterns and their internal structure.

At temperatures below  $0.5 T_m$  the structure evolves with increasing strain from cells with tangled boundaries into cells with boundaries consisting of wavy parallel dislocations and finally dislocation cell boundaries consisting of arrays of straight parallel dislocations. The cell structure is more defined (the DBs are sharper, e.g. the dislocation density inside them is higher) and the cell size is 0.5–1  $\mu\text{m}$ . Increasing strain causes sharpening of the DBs and

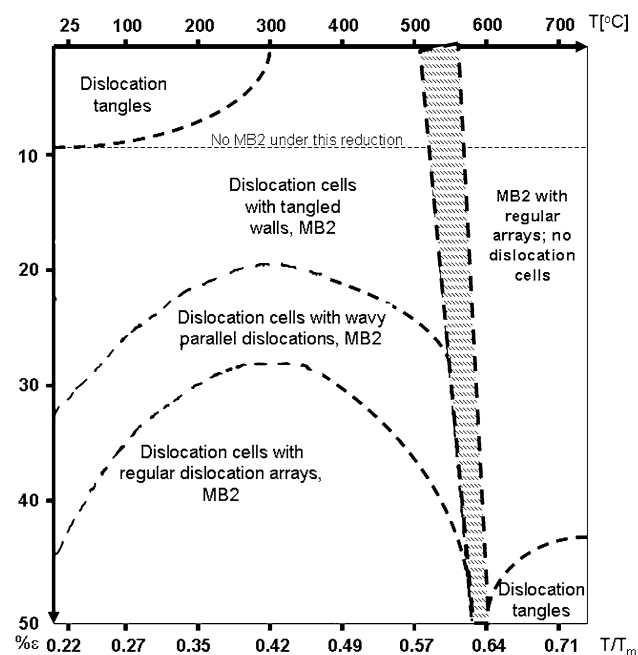
ordering of the dislocations into piecewise arrays. The cellular structure observed is of type 2 boundaries; the grains are subdivided by ordinary equiaxed dislocation cells. The MB2 were observed above 10% strain and their density increased with increasing strain, as was previously reported [20–22]. It seems that since the mobility of dislocations in copper is lower than that of aluminum (lower SFE) another pattern, MB2, is formed as an additional mechanism to compensate for the geometrical constraints.

Increasing the deformation temperature, i.e. the homologous temperature, caused a similar effect of sharpening of the cell walls and an increase in the MB2 density.

In copper, higher deformation temperatures lower the strain required for the dislocations to rearrange themselves into ordered arrays. For example, at room temperature, 50% strain was necessary for the dislocations to rearrange; however, at 350 °C only 45% strain was required. The effect of temperature is less significant than that of strain since the SFE of copper is relatively low.

Above 500 °C recrystallization was evident. At the higher temperatures ( $\geq 500$  °C) a new class of wider MB2 is observed. Their width is one order of magnitude higher than that of those observed at the lower temperatures. Clusters of both types of MB2 were more abundant at elevated temperatures and high strains. Above 500 °C, it seems that there is a transition temperature, at which a cell structure is no longer formed and the dislocations accumulate in the MB2 walls. This implies that the cellular structure was recovered into new grains. Residual dislocations are observed and at 50% strain they form tangles at the vicinity of the MB2.

The MB2 walls consist of parallel arrays of dislocations (Fig. 8) and each wall consists of more than one array, similar to the internal structure of DBs in aluminum and copper. This structure was observed in the MB2 at all the temperatures.



**Fig. 7** Microstructural map summarizing the different dislocation patterns observed in compressed copper at various temperatures and strains



**Fig. 8** TEM bright field image of a MB2's wall formed following 50% strain at 650 °C (arrowed). The dislocations form arrays of parallel dislocations

No “loosely knitted” dislocations structure in the MB2's walls, such as reported by Leffers et al. [17], was observed.

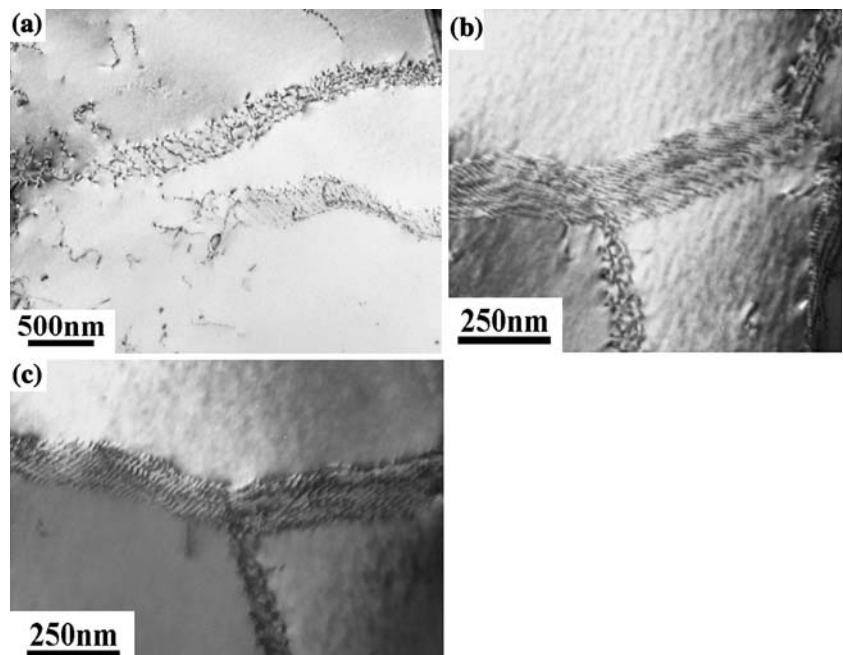
It seems that dislocation cells and MB2 are two competitive dislocation patterns, and formation of MB2 is preferred when the mobility, provided by temperature, and opportunity for rearrangements of dislocations, provided

by strain, are high. This is manifested in the increasing density of MB2 with higher strain and temperature. Recovery phenomena seems to eliminate the cellular structure, however, the formation of MB2 is less susceptible to temperature. The increase in the MB1 width with temperature [29] and our observation of the wider MB2 above 500 °C probably result from a recovery process.

It is interesting to compare the detailed structure of DBs in copper with those formed in aluminum. To this end we examined [31] samples of aluminum compressed at room temperature (Fig. 9). In copper, DBs consisting of undulating dislocations were observed following medium strains (20%) at a relatively high homologous temperature ( $0.45 T_m$ ) (Fig 3b). These patterns were similar to those observed in aluminum following room temperature deformation ( $0.3 T_m$ ) to relatively low strains (Fig. 9a).

Higher strain (50%) at  $0.45 T_m$  in copper produced sharp, ordered DBs (Fig. 4c) which were similar to those observed following medium strains (>15%) in aluminum deformed at room temperature. Figure 9b,c is an example of the same DB formed following 30% strain in aluminum in two different tilt angles. The DB consists of two arrays of parallel dislocations, similar to that observed in copper following 50% at  $0.45 T_m$ . It appears that in order to achieve the same internal structure as was observed in aluminum there has to be interplay of strain and temperature. A higher homologous temperature (increasing the dislocation mobility) and a higher strain (giving opportunity for the dislocations to form ordered structures) are required to form the same internal structures in copper. Increasing the deformation temperature compensates for

**Fig. 9** TEM bright field image of DBs in compressed pure polycrystalline aluminum [30]: (a) 10% strain at room temperature. The dislocations are wavy. (b) 30% strain at room temperature. The DB consists of an array of parallel dislocation. (c) 30% strain at room temperature. Same DB as in (b) at a different tilt angle revealing a second burgers vector



the higher melting temperature of copper, but due to the lower SFE of copper an additional increase in strain is required. Higher temperatures (above  $0.5 T_m$ ) cause recovery of the microstructure. This is manifested in the disappearance of the cellular structure.

## Conclusions

The evolution of the microstructure with increasing strain at various temperatures was studied in pure polycrystalline copper following compression and a microstructural map was produced. It was shown that up to 500 °C the effect of temperature resembles the effect of strain. With increasing strain and temperature the cell structure becomes defined and the density of MB2 increases. The detailed structure of the DBs evolves from tangled dislocations into arrays of parallel dislocations with increasing strain and temperature. Interplay between strain and temperature enables the dislocations in copper to form a detailed structure of the DBs similar to the one formed in aluminum at room temperature deformation.

The detailed structure of MB2 was found to consist of arrays of parallel dislocations. At elevated temperatures (above  $0.5 T_m$ ) recrystallization, recovery and grain growth occurs and the microstructure consists solely of MB2. At these temperatures a second type of MB2 is formed, one order of magnitude wider than the one observed at the lower temperatures.

## References

- Hansen N, Jensen DJ (1999) *Phil Trans R Soc Lond A* 357:1447
- Bay B, Hansen N, Hughes DA, Kuhlmann-Wilsdorf D (1992) *Acta Metall Mater* 40:205
- Hansen N, Hughes DA (1995) *Phys Stat Sol (b)* 149:155
- Hughes DA (2002) *Script Mater* 47:697
- Hughes DA, Hansen N (1997) *Acta Mater* 45:3871
- Swann PR (1963) In: Thomas G, Washburn J (eds) *Electron microscopy and strength of crystals*. Interscience, NY p 131
- Hansen N (1992) *Script Metall Mater* 27:1447
- Landau P, Shneck RZ, Makov G, Venkert A (2007) In: Balk TJ, Minor A, Porter A, Plitzko J (eds) *Electron microscopy across hard and soft materials*, *Mat. Res. Soc. Symp. Proc.* 982E. Warrendale, PA, 0982-KK07-04
- Kuhlmann-Wilsdorf D (1989) *Mat Sci Eng A* 131:1
- Godfrey A, Jensen DJ, Hansen N (1998) *Acta Mater* 46:835
- Caballero V, Varma SK (1999) *J Mat Sci* 34:461
- Wagner P, Engler O, Lucke K (1995) *Acta Metall Mater* 43:3799
- Hansen N (1990) *Mat Sci Tech* 6:1039
- Huang X (1998) *Scrip Mater* 38:1697
- Huang X, Borrego A, Pantleon W (2001) *Mat Sci Eng A* 319–321:237
- Ananthan VS, Leffers T, Hansen N (1991) *Mat Sci Tech* 7:1069
- Leffers T, Anathan VS, Christofferson H (2001) *Mat Sci Eng A* 319:148
- Anathan VS, Leffers T, Hansen N (1991) *Scrip Metall Mater* 25:137
- Gonzalez B, Murr LE, Valerio OL, Esquivel EV, Lopez H (2003) *Mat Char* 49:359
- Malin AS, Hatherly M (1979) *Met Sci* 13:463
- Huang JC, Gray III GT (1989) *Acta Metall* 37:3335
- Hatherly M (1992) *Scrip Metall Mater* 27:1453
- Korbel A, Martin P (1986) *Acta metal* 34:1905
- Kim YW, Bourell DL (1988) *Metall Trans A* 19:2041
- Kohlhoff GD, Malin AS, Lucke K, Hatherly M (1988) *Acta Metall* 36:2841
- Malin AS, Huber J, Hatherly M (1981) *Z Metallk* 72:310
- Bassim MN, Liu CD (1993) *Mat Sci Eng A* 164:170
- Jackson PJ (1983) *Scrip Metall* 17:199
- Park NK, Parker BA (1989) *Mat Sci Eng A* 113:431
- Gan J, Vetrano JS, Khaleel MA (2002) *J Eng Mat Tech* 124:297
- Landau P (2005) *Microstructure evolution of deformed fcc metals*. M.Sc thesis, Ben-Gurion University, Israel
- Caillard D, Martin JL (1982) *Acta Metall* 30:437
- Anongba PNB, Bonneville J, Martin JL (1993) *Acta Metall Mater* 41:2897
- Anongba PNB, Bonneville J, Martin JL (1993) *Acta Metall Mater* 41:2907
- Straub S, Blum W, Maier HJ, Unger T, Borbely A, Renner H (1996) *Acta Mater* 44:4337
- Belyakov A, Sakai T, Miura H, Tsuzaki K (2001) *Phil Mag A* 81:2629