JOURNAL OF MATERIALS SCIENCE 26 (1991) 1734–1740

Recrystallization in β brass

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Recrystallization has been studied in β brass in order to evaluate the influence of order on the recrystallization process. Recrystallization takes place by the formation of new grains at shear band – grain boundary intersections followed by grain growth and coalescence. Recrystallization occurs most readily at temperatures near the critical ordering temperature where the alloy has partial or short-range order and this is because both new grain formation and grain growth are fastest. Dislocation recovery processes are more difficult within the partially ordered materials and a larger strain energy is retained to drive recrystallization.

1. Introduction

Recrystallization in ordered alloys has received limited attention and the role of order on influencing recrystallization remains uncertain. Initial studies [1–3] suggested that recrystallization did not occur on heating a deformed, ordered alloy, but later studies [4–9] have shown that recrystallization can occur, albeit more slowly when in the ordered state. Essentially two families of hypotheses have been developed to explain the slower kinetics of the ordered state – that a recrystallizing grain boundary will have a lower mobility within an ordered material, or that interactions between dislocations and ordered defects such as antiphase boundaries (APBs) affect dislocation recovery and inhibit the recrystallization process.

The low mobility of a boundary within an ordered material may be a lower intrinsic mobility because of the more difficult atomic motion within the ordered lattice [10], as probably observed by Davies and Stoloff [5] when studying grain growth in a Fe–Co alloy. The low boundary mobility may also be caused by a more complex boundary structure for the ordered state. For example, Hutchinson *et al.* [6] suggest a change in boundary structure from being a high angle boundary in the disordered alloy to a low angle boundary in the ordered alloy. This conclusion was based on the different orientation textures produced when recrystallizing the ordered and disordered materials. The same explanation for low boundary mobility in the ordered state was proposed by Gottstein *et al.* [9] even though their texture data was seen to be incompatible with the results of Hutchinson *et al.* [6]. Some indication of the difficulty of grain boundary motion in an ordered alloy is perhaps given by the ordered faults seen trailing behind the advancing grain boundaries [6, 8, 11].

Interactions between dislocation recovery and the ordered structure or the APB faults have been seen on many occasions. Corey and Potter [12] observed that dislocation recovery occurred easily in deformed Ni_3Al as ordering took place. Davies [13] found that the ordering kinetics could be accelerated by cold work, while Ward and Mikkola [14] in a detailed examination of deformed Cu_3Au found accelerated initial domain growth kinetics, as ordering and dislocation recovery occurred, and later slower domain growth kinetics. Contrary to these results, Hutchinson *et al.* [6] found that prior cold work hardly affected ordering or domain growth. In addition to these studies where deformation can lead to changes in the rate of ordering, several studies have also shown that order can affect dislocation mobility, and hence recovery and recrystallization [15, 16]. In these studies partial order has been shown to inhibit recovery processes, such as polygonization, and hence to slow recrystallization. Greenberg [17, 18] has attempted to classify the affects of ordering on recrystallization in terms of the relative kinetics of the two processes, based on the principle of rapid ordering tending to freeze the dislocation structure and inhibit the rearrangements necessary for recrystallization. Cahn [19] has suggested that it may be the fine domain structure present during the early stages of ordering that hinders the nucleation of recrystallization by inhibiting the dislocation rearrangements near or at grain boundaries and slowing grain boundary migration.

In the present study, the recrystallization kinetics have been examined in a disordering alloy such that the influence of partial or complete order can be studied by comparison with the behaviour of the disordered state. For alloy systems where the order–disorder critical temperature is low, studies of recrystallization in the ordered state are restricted to such low temperatures that only recovery occurs, not recrystallization, and this may lead to anomalous apparent recrystallization behaviour. For example in the case of Cu_3Au the critical ordering temperature is about 390 °C and annealing studies at low temperature show extensive dislocation recovery before recrystallization [6]. The present study has been carried out on β brass which has a higher critical ordering temperature, about 470 °C [20], and recrystallization kinetics remain reasonably fast both above and below the critical temperature. Detailed metallographic studies are carried out to examine nucleation and

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grain growth kinetics during annealing to recrystallize, and the changes in dislocation structures within the unrecrystallized parts are examined to see the extent of recovery occurring.

2. Experimental details

The alloy was supplied in bar form with a measured composition of 52% copper and 48% zinc (at %). The initial grain size was about 500 μm following a recrystallization heat treatment at 480 °C. The material was found to be too brittle to deform by drawing techniques and hence cylindrical samples of 2:1 aspect ratio were deformed 20% in compression. Heat treatments on small pieces were carried out in muffle furnaces under argon atmosphere or, for short anneals, in a salt bath. Comparative anneals under both annealing conditions showed no differences.

Optical metallography was performed on polished and etched samples. The microstructure was uniform through the sample cross-sections apart from a narrow surface region of fine grains which was not considered during the study. Quantitative metallography was carried out manually using standard techniques: the surface fraction of recrystallized material was measured using a point counting technique; grain sizes were measured to give both an average and a maximum size of recrystallized grain; the number of new grains per unit volume was estimated as the measured number per unit surface divided by the average grain size.

Transmission electron microscopy was carried out using a Philips CM 12 instrument on thin foils prepared by twin-jet electropolishing using a 10% nitric acid in methanol solution at about 15 V and − 20 °C. In addition, estimates of the extent of recovery occurring were made by hardness measurements on the unrecrystallized parts of each sample.

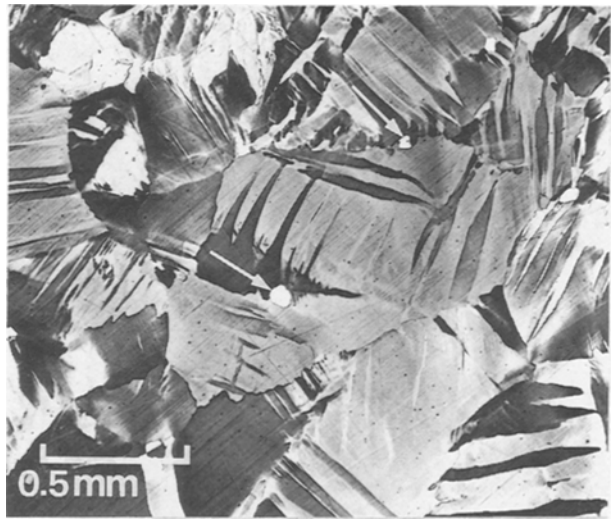


Figure 1 Optical micrograph showing deformation bands within grains and a number of small, new grains forming at the beginning of recrystallization (arrowed). Annealed 5 s at 480 °C.

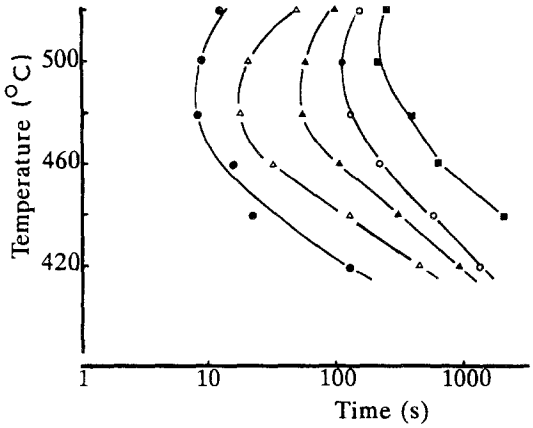


Figure 2 Recrystallization kinetics of β brass deformed 20%. Numbers on curves represent the fraction recrystallized. (● 0.05, △ 0.2, ▲ 0.4, ○ 0.6, ■ 0.8).

3. Results

Large numbers of deformation bands are produced during straining, and on annealing new grains form initially on such bands, particularly at intersections with grain boundaries, see Fig. 1. Such nucleation occurs rapidly initially and later slows as though suitable nucleation sites have become saturated. During this first stage the individual new grains grow until impingement occurs, and then a slower grain coarsening process takes over.

The kinetics of recrystallization are shown in Fig. 2. Recrystallization is fastest at 480 to 500 °C, over the temperature range where this alloy has just disordered. The grain growth kinetics at each temperature are shown in Fig. 3. The evolution of grain size may be separated into three stages, even though the three stages are not clearly distinguished at all temperatures in view of the difficulties of examining very short recrystallization treatments and the limited data at long times. In the first stage the grain size grows linearly with time – this is the stage of formation of isolated grains. In the second stage these small grains meet and grow by coalescence such that the grain size increases approximately with $(\text{time})^{1/2}$. Towards the end of recrystallization, grain growth appears to slow even more. At this stage there is some difficulty in distinguishing between the growing, new grains and the remaining, recovered but unrecrystallized material. The nucleation rate of new grains has been measured during the first stage indicated by Fig. 3 as the number of new grains per unit volume divided by the annealing time. The results are summarized in Table I. The data are subject to considerable uncertainty because of the difficulty of detecting very small grains at the short times considered. The variation of nucleation rate with temperature is slight, showing the highest value at 500 °C.

The kinetics of grain growth and coarsening, from the first two stages in Fig. 3, may be related to the annealing temperature by an Arrhenius relationship as shown in Fig. 4. When in the ordered state, below ~ 470 °C, the grain growth rate fits the Arrhenius relationship with an activation energy of 145 kJ mol^{-1}. This value is very close to that of diffusion of copper in ordered β brass [21]. When in

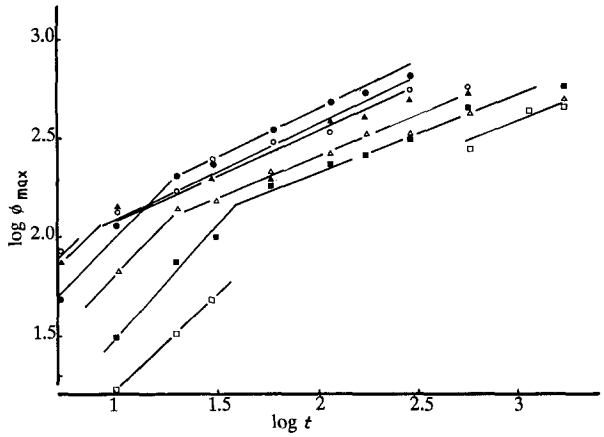


Figure 3 Size (ϕ) of recrystallized grains during annealing. An initial stage of isolated grain growth and a second stage of grain coarsening can clearly be distinguished. At very long times grain growth slows further. (□ 420 °C, ■ 440 °C, △ 460 °C, ▲ 480 °C, ○ 500 °C, ● 520 °C).

TABLE I Nucleation rate of new grains during the initial stage of annealing

Temperature (°C)	Nucleation rate ($m^3 s^{-1}$)
420	15 ± 5
440	30 ± 5
460	23 ± 5
480	35 ± 10
500	50 ± 10
520	45 ± 10

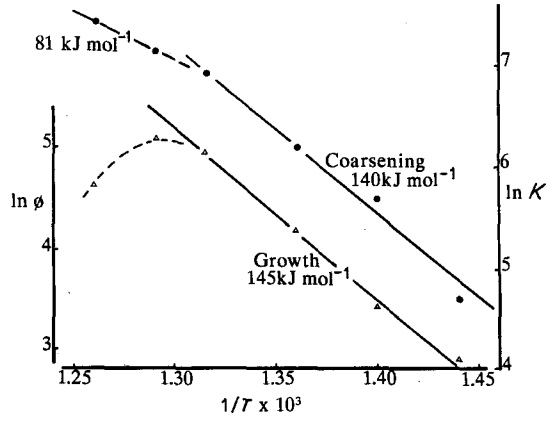


Figure 4 Arrhenius plots of grain growth rate ($\dot{\phi}$) and coalescence rate ($K = d\phi^2/dt$) for ordered (< 470 °C) and disordered (> 470 °C) β brass.

the disordered state, above 470 °C, the grain growth rate decreases with increasing temperature indicating that some other process controls behaviour. The kinetics of coarsening also fit the Arrhenius relationship with activation energies of 140 and 81 kJ mol^{-1} in the ordered and disordered states, respectively. The latter value is close to that for the activation energy of diffusion in disordered β brass [21].

The degree of dislocation recovery within the unrecrystallized parts is estimated from hardness measurements made in these parts, as shown in Fig. 5. A large fall in hardness occurs within 5 to 10 sec only,

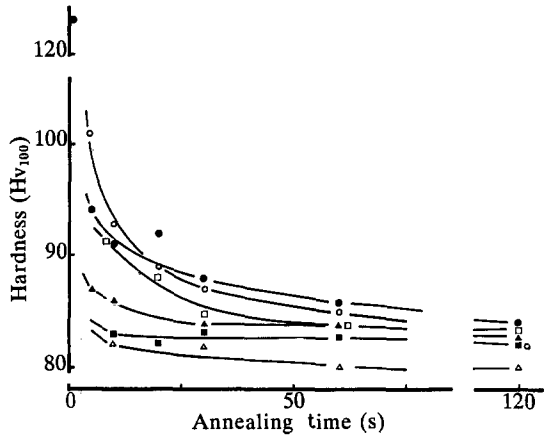


Figure 5 Hardness of unrecrystallized regions on annealing to induce recrystallization (□ 420 °C, ■ 440 °C, △ 460 °C, ▲ 480 °C, ○ 500 °C, ● 520 °C).

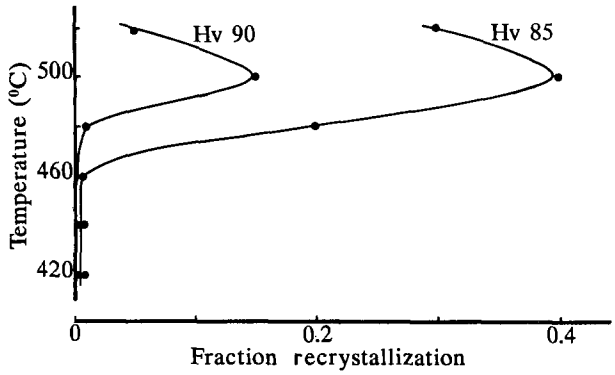


Figure 6 Fraction of recrystallization corresponding to the time when a given degree of hardness recovery has occurred.

that is at the very earliest stages of recrystallization. It is clearly seen, however, that the rate of hardness recovery varies significantly with the heat treatment temperature and is much faster at 440 to 480 °C than at 500 to 520 °C and at 420 °C. The much slower rate of recovery around 500 °C is clearly seen in Fig. 6, which shows the extent of recrystallization by the time a given hardness recovery is achieved. At high and low temperatures (> 520 °C and < 480 °C) hardness recovery occurs essentially before recrystallization has started; at 480 to 520 °C recovery is so slow that significant recrystallization occurs before the hardness has fallen.

The following micrographs illustrate the changes in dislocation arrangements within the unrecrystallized parts during annealing. Micrographs are shown only for samples annealed at 440 and 500 °C, which are typical of the ordered and disordered states just above the critical temperature, respectively. Figs 7 to 10 illustrate microstructures within ordered samples after recrystallization to about 5% (Figs 7 and 8), and 70% (Figs 9 and 10). At the early stages of recrystallization, dislocation recovery to a loose cell structure has already taken place in the ordered material. All the dislocations are paired as superdislocation pairs. After annealing for longer times, a well formed subgrain structure is seen with subgrain boundaries composed of superdislocations. Within the recrystallized grains

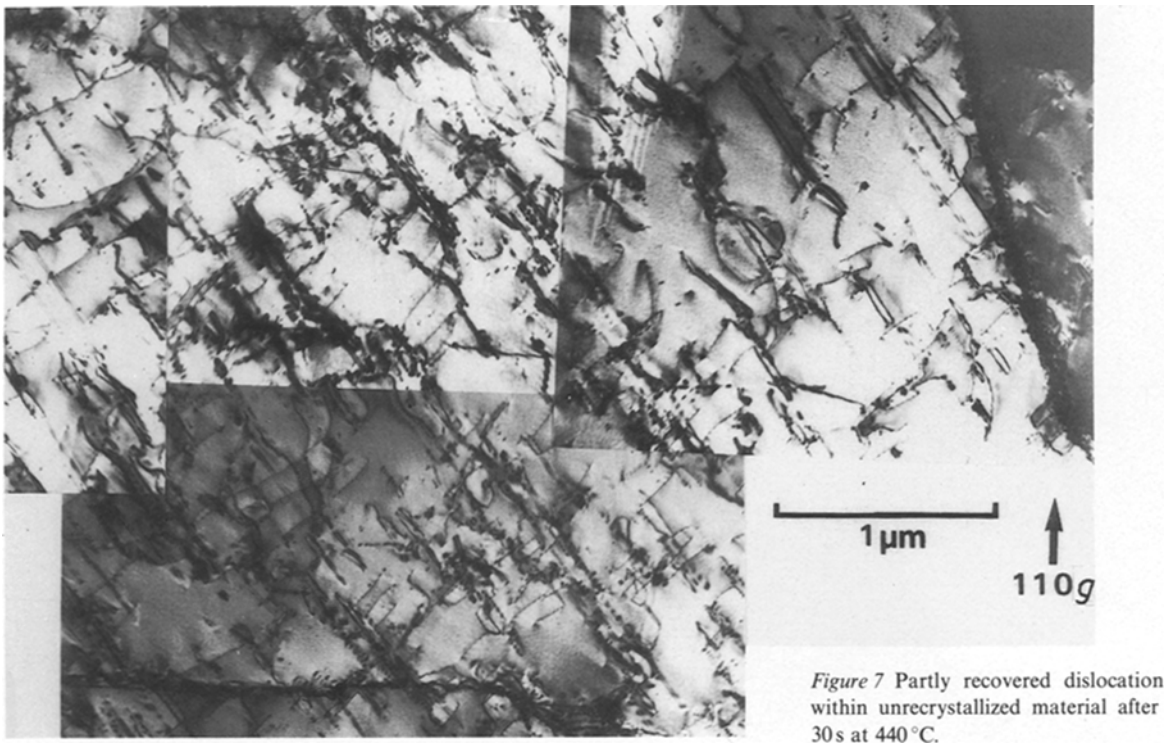


Figure 7 Partly recovered dislocation within unrecrystallized material after 30 s at 440 °C.

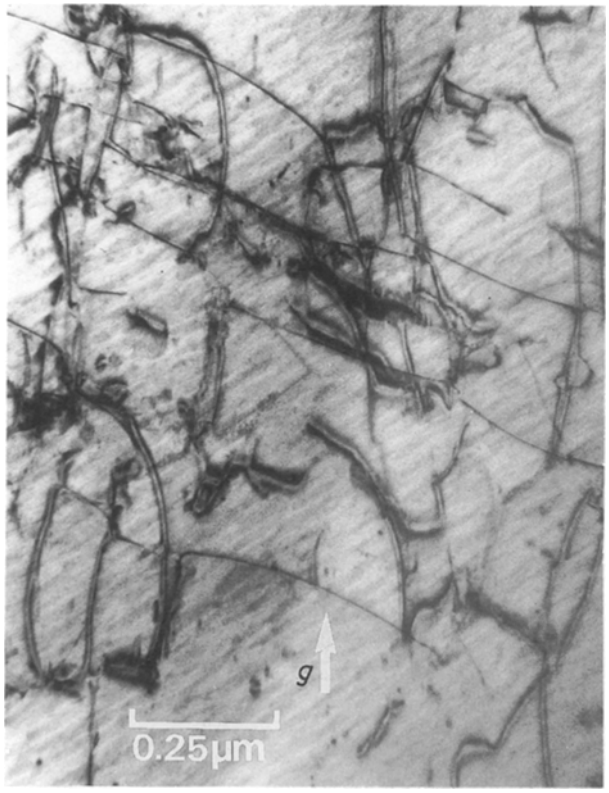


Figure 8 Superdislocations remaining within unrecrystallized part after annealing 30 s at 440 °C.

there are no traces of an antiphase domain boundary network, perhaps because domain growth is fast for this alloy, and there are essentially no dislocations or other defects apart from a few straight antiphase boundary faults attached to dislocations; Fig. 11.

Figs 12 to 16 illustrate typical dislocation arrangements seen within the unrecrystallized parts of material heated at 500 °C to recrystallize about 5% (Fig. 12) and 60% (Figs 13 to 16). The arrangements seen are typical also of disordered materials heated at 480 and 520 °C. Annealing to recrystallize 5 and 60%, Figs 12 and 13, leads to only partial dislocation recovery and only traces of cell formation. Many of the dislocations are present as dipoles, see Fig. 14, which annihilate slowly by forming dislocation loops which eventually disappear, see Fig. 15. Even within the disordered material, most of the dislocations are present as pairs, see Fig. 16. These are coupled dislocations having the same Burgers vectors and are produced because of the considerable short range order remaining at the annealing temperature, 500 °C, slightly above the critical temperature for the disappearance of long range order.

4. Discussion and summary

Of particular interest at the present time is a comparison of the mechanisms and rates of recrystallization in ordered and in disordered materials. The most significant results obtained here are of fastest recrystallization kinetics, grain nucleation and grain growth just above the order–disorder critical temperature, 480 to 500 °C. Secondly, no abrupt changes in kinetics are seen to occur at the critical temperature.

The nucleation rate and growth rate under a given condition may be related to the dislocation mobility for the particular process controlling relaxation or recrystallization and to the driving force. The dislocation evidence here shows clearly that easy recovery takes place at low temperatures in the ordered state, leaving less driving force, whilst at temperatures near or above the critical temperature for disordering recovery is much slower. This is believed to be caused by short range order in the disordered state, perhaps also a high degree of partial disorder at temperatures just

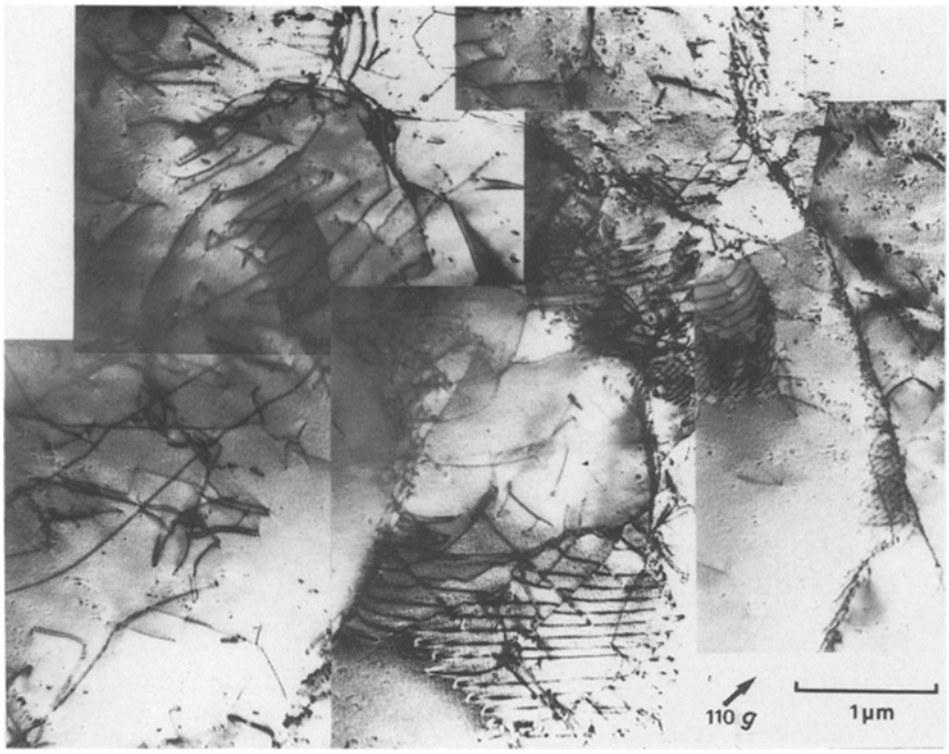


Figure 9 Well recovered subgrain structure within unrecrystallized part after annealing 30 min at 440 °C.

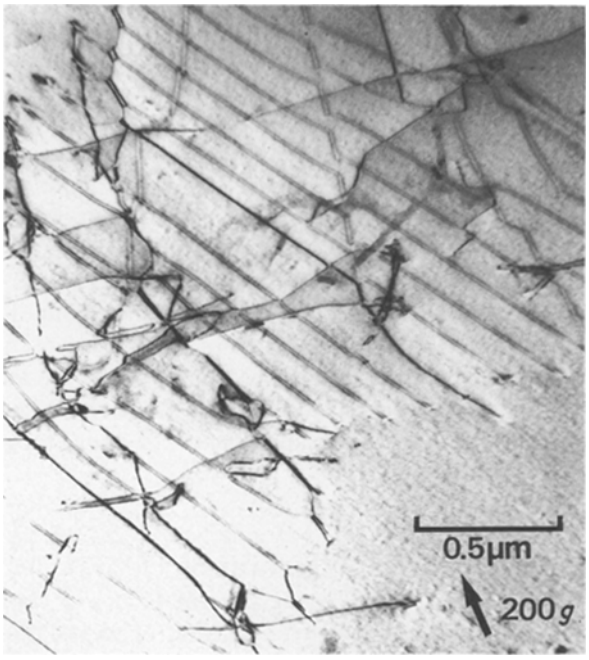
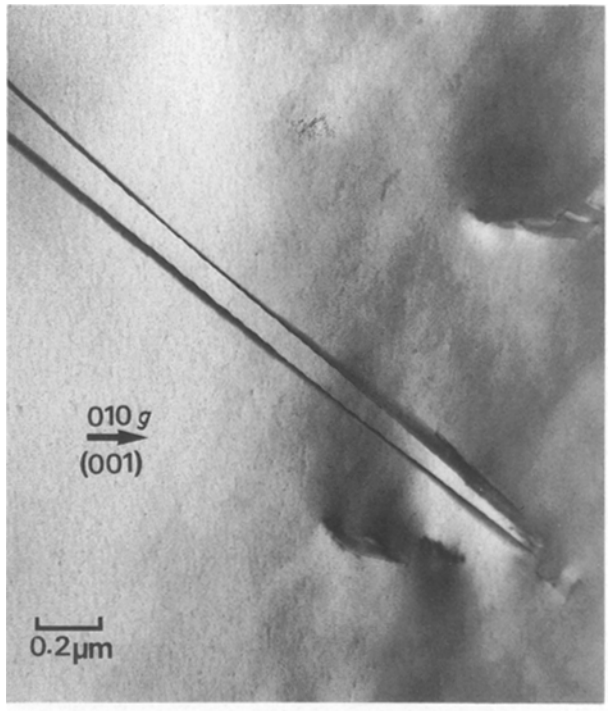


Figure 10 Well formed subgrain boundary composed of superdislocations after annealing 30 min at 440 °C.



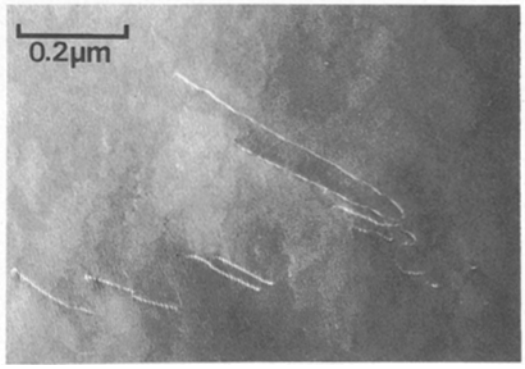


Figure 11 Stacking faults within recrystallized region after annealing 30 min at 440 °C.

below the critical ordering temperature, which makes dislocation motion more difficult. Evidence for the existence of such short range order is given by the coupling of dislocations even when annealed in the nominally disordered state and by the diffuse background seen in the electron diffraction patterns for these materials. The reason for fast recrystallization at 480 to 500 °C is then clear: because recovery is slow, a large driving force remains for easy grain formation (see Table I) and for fast grain growth. At such high temperatures grain coarsening processes are also rapid. At lower temperatures, more perfect order allows

Figure 12 Partially recovered dislocation structure within the unrecrystallized parts after annealing 10 s at 500 °C to recrystallized 5%.

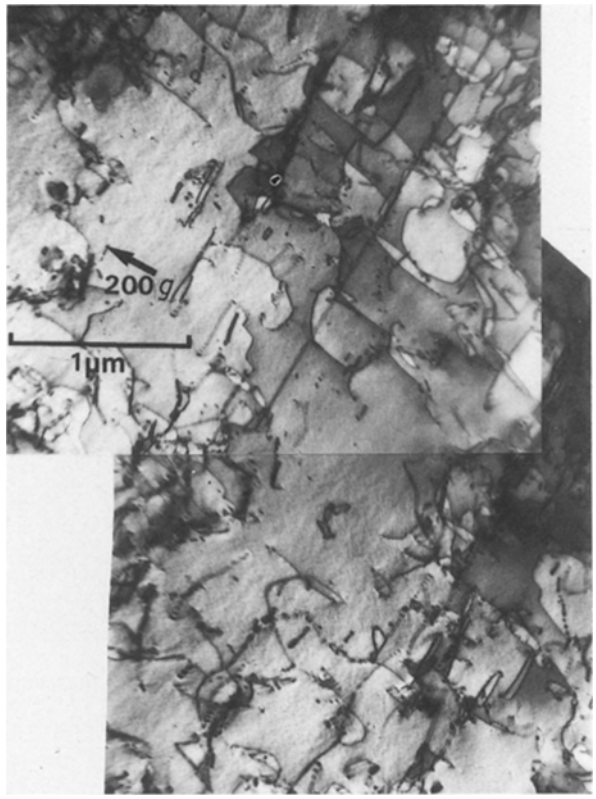


Figure 13 Partially recovered dislocation structure within the unrecrystallized parts after annealing 120 s at 500 °C to recrystallize 60%.

easier dislocation recovery such that the driving force for recrystallization falls: both because of this and because of the lower degree of thermal activation, the nucleation rate, the growth rate and also the grain coarsening rate decrease. At the highest temperatures considered, dislocation recovery may again become rapid as short range order is lost. Nucleation and growth rates then fall (Table I and Fig. 4). The rate of grain coarsening, a process driven by the surface energy of the growing grains, continues to increase, Fig. 4.

Of particular interest is the continuity in the growth rate and coarsening rate about the critical ordering temperature. Apart from a change in the activation

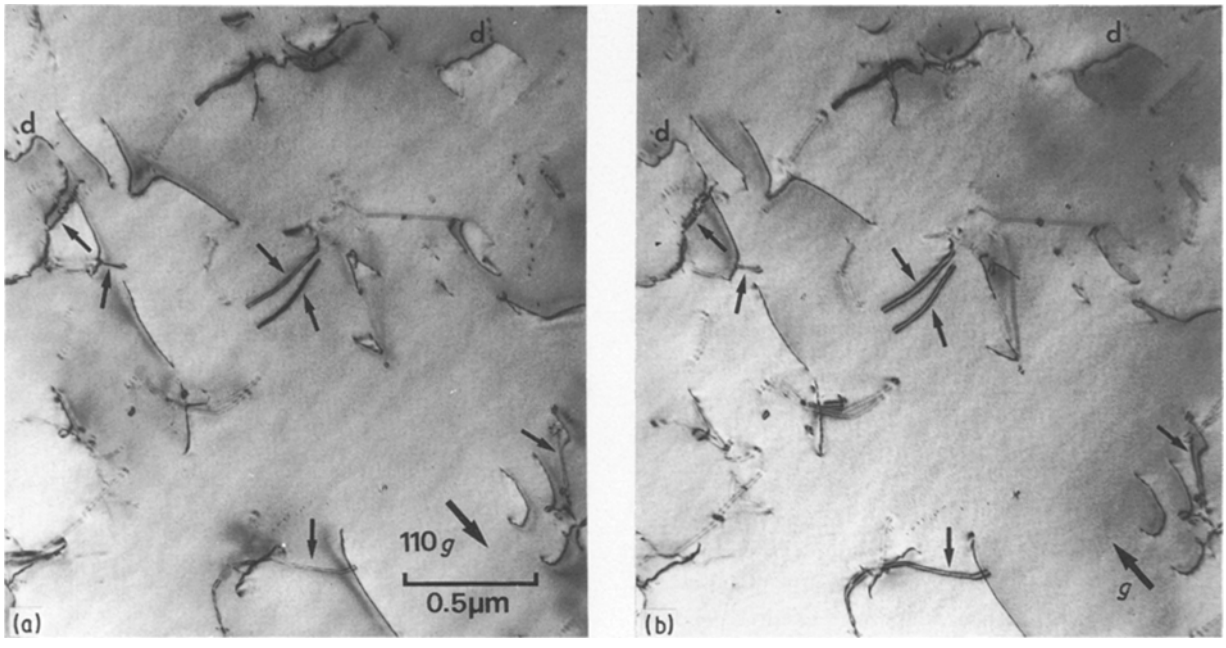


Figure 14 Pair of micrographs using opposite sign g vector to illustrate the large number of dipoles (arrowed). Annealed 120 s at 500 °C.

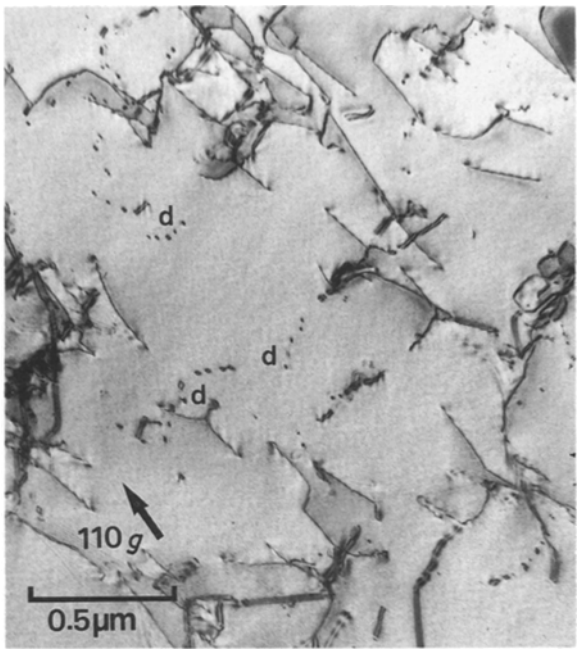


Figure 15 Debris (d) remaining after dipole annihilation in material annealed 120 s at 500 °C.

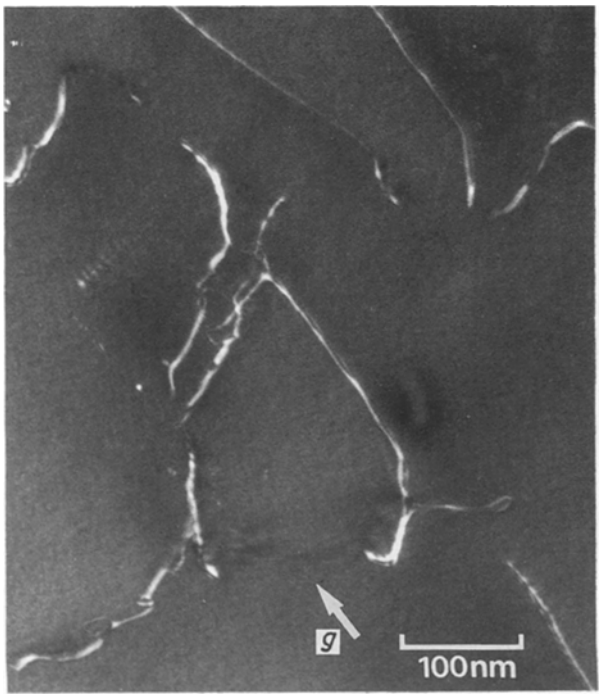


Figure 16 Paired dislocations of same Burgers vector in material annealed 120 s at 500 °C.

energy of the controlling diffusional process there is no sudden change in grain coarsening rate, see Fig. 4. This implies that the grain boundary structure and processes occurring at the grain boundary, must be essentially the same for both the ordered and the disordered state.

Finally, in view of the similarity of recrystallization kinetics in the ordered and disordered states seen here, it is of interest to reconsider previous results showing different material behaviour when ordered or disordered. Firstly, it should be noted that the slower diffusivity, and larger activation energy of diffusion of the ordered state will lead to generally slower recrystallization kinetics when ordered, as commonly observed. It is also possible that the long range ordered state, in the absence of significant additional short range order, may induce fast dislocation recovery–as observed by Ward and Mikkola [14] who found that initial domain growth kinetics were fast, presumably as dislocations moved in directions to reduce antiphase domain boundary surface and energy. A reduced driving force for recrystallization, as a result of enhanced dislocation recovery, would certainly slow the recrystallization kinetics and could even lead to situations where the dislocation structure may be sufficiently stabilized by extensive recovery that no recrystallization occurs.

As a final comment, it should be noted that β brass, as an alloy capable of being thermally disordered, may in fact always have a region of disorder associated with the grain boundaries, for example similar to that postulated for $Ni_3Al + B$ [22]. It is possible that ordered intermetallics which do not possess such disordered grain boundaries may show different recrystallization characteristics.

References

1. M. M. BORODKINA, E. I. DETALF and Y. D. SELISSKII, *Phys. Met. Metallog.* **7** (1959) 50.
2. Y. P. SELISSKII and M. N. TOLOCHKO, *Phys. Met. Metallog.* **13** (1962) 98.
3. A. E. VIDOZ, D. P. LAZAREVIC and R. W. CAHN, *Acta Metall.* **11** (1963) 17.
4. A. A. GOLDENBERG and Y. P. SELISSKII, *Phys. Met. Metallog.* **15** (1963) 64.
5. R. G. DAVIES and N. S. STOLOFF, *Trans. Met. Soc. A.I.M.E.* **236** (1966) 1605.
6. W. B. HUTCHINSON, F. M. C. BESAG and C. V. HONESS, *Acta Metall.* **21** (1973) 1685.
7. G. R. HAFF and E. M. SCHULSON, *Met. Trans.* **13A** (1982) 1563.
8. I. BAKER, D. V. VIENS and E. M. SCHULSON, *J. Mater. Sci.* **19** (1984) 1799.
9. G. GOTTSTEIN, P. NAGPAL and W. KIM, *Mater. Sci. Eng.* **A108** (1989) 165.
10. A. A. GOLDENBERG and Y. P. SELISSKII, *Fiz. Metal. Metalloved.* **15** (1963) 717.
11. Y. CALVAYRAC and M. FAYARD, *Acta Metall.* **14** (1966) 783.
12. C. L. COREY and D. I. POTTER, *J. Appl. Phys.* **38** (1967) 3894.
13. R. G. DAVIES, *J. Phys. Chem. Solids* **24** (1963) 985.
14. A. L. WARD and D. E. MIKKOLA, *Met. Trans.* **3A** (1972) 1479.
15. M. FELLER-KNEIPMEIER and F. RUCKERT, *Z. Metallkde.* **66** (1975) 427.
16. R. A. BUCKLEY, *Met. Sci.* **13** (1979) 67.
17. B. A. GREENBERG and YU. N. GORNOSTIREV, *Scripta Metall.* **19** (1985) 1391.
18. *Idem. ibid.* **19** (1985) 1397.
19. R. W. CAHN, "Recrystallization, Grain Growth, and Textures", edited by H. Margolin, (American Society of Metals, Metals Park, OH, 1966) p. 99.
20. M. M. SHEA and N. S. STOLOFF, *Met. Trans.* **5** (1974) 577.
21. A. B. KUPER, D. LAZARUS, J. R. MANNING and C. T. TOMIZUKA, *Phys. Rev.* **104** (1956) 1536.
22. R. A. D. MACKENZIE and S. L. SASS, *Scripta Metall.* **22** (1988) 1807.

Received 9 October 1989
and accepted 24 April 1990