



HAL
open science

DIFFUSION INDUCED GRAIN BOUNDARY MIGRATION IN ASYMMETRICAL TILT GRAIN BOUNDARIES

A. King, G. Dixit

► **To cite this version:**

A. King, G. Dixit. DIFFUSION INDUCED GRAIN BOUNDARY MIGRATION IN ASYMMETRICAL TILT GRAIN BOUNDARIES. Journal de Physique Colloques, 1990, 51 (C1), pp.C1-545-C1-550. 10.1051/jphyscol:1990185 . jpa-00230353

HAL Id: jpa-00230353

<https://hal.science/jpa-00230353>

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

DIFFUSION INDUCED GRAIN BOUNDARY MIGRATION IN ASYMMETRICAL TILT GRAIN BOUNDARIES

A.H. KING and G. DIXIT

Department of Materials Science and Engineering, State University of New York at Stony Brook, Stony Brook NY 11794-2275, U.S.A.

Abstract – The phenomenology of diffusion induced grain boundary migration (DIGM) is studied as a function of misorientation angle in a series of asymmetrical tilt grain boundaries with [100] and [110] rotation axes. The diffusion of zinc into copper is investigated under constant conditions for all of the specimens. Interesting comparisons are drawn with previous results on symmetrical boundaries, and with the various theories of DIGM.

1 – INTRODUCTION

Diffusion induced grain boundary migration has been studied widely for a little over ten years now, but there remains a dearth of systematic data about the phenomenon [1]. Most experimenters report either observations of the phenomenon in new alloy systems, or fairly detailed studies of it in a single grain boundary. There are broadly only two exceptions to these kinds of study. First, Yoon and co-workers [2,3] have set out systematically to investigate the effect of lattice misfit upon the progress of liquid film migration, and coincidentally obtained information about DIGM. Second, Chen and King [4] have studied DIGM as a function of misorientation in a series of symmetrical tilt grain boundaries. It has been pointed out [5] that, so far, these experiments have been somewhat incompatible with each other and not suitable for comparison. In order to rectify these problems, we have undertaken an extension of the work by Chen and King, to include asymmetrical tilt grain boundaries, in order to provide data that can usefully be compared with that from Yoon's group, and also with the established theories of DIGM [6,7].

2 – THEORETICAL CONSIDERATIONS

Two major theories have been proposed to explain the phenomenon of DIGM. These are the "coherency strain" mechanism [6] and the "dislocation climb" mechanism [7]. In the coherency strain mechanism, the migration of the boundary is postulated to occur because of the asymmetrical build-up of strain as solute leaks out of the boundary into the surrounding lattice. In the limiting case, or where the strain is completely relieved on one side of the boundary by plastic deformation, a strained layer exists on one side of the boundary only. Under conditions of asymmetrical strain, the energy of the system can be reduced if the grain boundary migrates through the strained layer. The velocity of the boundary is given by

$$v = \frac{D_G V_m}{\delta RT} [Y(\mathbf{n}_1) - Y(\mathbf{n}_2)] \eta^2 (C_S - C_0)^2 \frac{C_G (1 - C_G)}{(C_G - C_S)^2} \quad (1)$$

where D_G is the diffusivity across the grain boundary, V_m is the molar volume, δ is the width of the grain boundary, $Y(\mathbf{n}_i)$ is an appropriate orientation dependent elastic modulus in grain i , η is the atomic misfit of the solute in the solvent lattice, C_S is the concentration of solute in the "leakage layer", C_0 is the original

concentration of the material, and C_G is the concentration in the grain boundary. The orientation dependence of Y is given, in terms of elastic constants, by

$$Y = \frac{(C_{11} + 2C_{12})}{2} \left[3 - \frac{C_{11} + 2C_{12}}{C_{11} + 2(2C_{44} - C_{11} + C_{12})(l^2 m^2 + m^2 n^2 + l^2 n^2)} \right] \quad (2)$$

Here, l , m and n are direction cosines between the relevant boundary normal and the standard crystallographic axes. From equations 1 and 2, we are able to calculate the variation of the expected velocity as the grain boundary plane changes.

In contrast with the coherency strain model, the dislocation climb model for DIGM postulates that grain boundary dislocations and their associated steps in the grain boundary plane, are induced to climb by the demand for point defects that arises from a grain boundary Kirkendall effect. Variations of the driving force are not considered in this model, but the grain boundary migration rate is found to vary as $h/b \sin \theta$, where h is the step height, b the Burgers vector magnitude, and θ is the angle between the Burgers vector and the boundary normal. It has been shown [8] that the possible values of h/b increase as the coincidence site density decreases. Thus as a general rule the migration rate should be higher for boundaries characterized by lower coincidence site densities.

3 - EXPERIMENTAL

All of our experiments are performed using copper bicrystals prepared by the Bridgman method. Experimental details are provided elsewhere [4]. The crystals are grown vertically in a graphite mold, under vacuum. The mold is designed with sharp protrusions on opposite sides, that form grooves in the bicrystal, in order to pin the grain boundary. Two series of bicrystals were grown, the first having a common [100] axis with one of the grains fixed with a (001) boundary plane. The second series of bicrystals had a common [110] axis with one of the crystals having a fixed (110) boundary plane. Individual specimens of thickness approximately 2mm were sliced from the bicrystals, and the surfaces were mechanically and chemically polished to remove all surface damage and thus prevent diffusion induced recrystallization from occurring [9]. Following this preparation, the specimens were encapsulated *in vacuo* with several grams of α -brass filings (30% Zn), then annealed at 400°C. After this diffusion anneal, the specimens were inspected using optical and scanning electron microscopy.

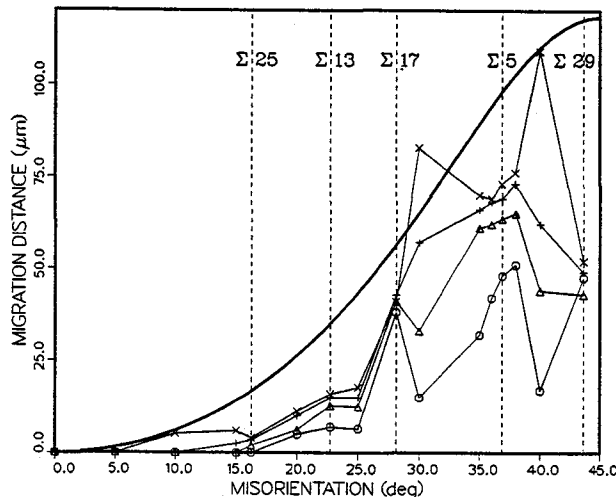


Figure 1. The variation of migration distance with misorientation for (100) asymmetrical tilt boundaries. The four experimental curves correspond to annealing times of 3, 6, 12 and 24 hours. The heavy, continuous curve represents the theoretical variation based upon the coherency strain driving force, and it is fitted to an arbitrary maximum.

4 - RESULTS

i) - (100) Boundaries.

The average migration distances for the (100) asymmetrical grain boundaries are shown in Fig.1, along with the expected variation of grain boundary velocity, derived from equation 1, fitted to an arbitrary maximum. It can be seen that the experimental data match the theoretical curve very well up to a misorientation of approximately 25°. At larger misorientations, however, there are large deviations from the

theoretical curve, especially for coincidence-related grain boundaries which appear to move rapidly at first, but quickly cease their migration. In every case the grain oriented with a (100) boundary plane was the grain that grew, which is as expected on the basis of the coherency strain mechanism. Figure 2 shows the average zinc concentration in the alloyed regions behind the moving boundaries after 24 hours. The coincidence boundaries $\Sigma 25$, $\Sigma 13$ and $\Sigma 5$ all exhibit notably high zinc concentrations, but $\Sigma 17$ and $\Sigma 29$ fail to show any special behavior. In previous work on symmetrical boundaries [4] $\Sigma 25$, $\Sigma 17$ and $\Sigma 5$ had high concentrations, but not $\Sigma 13$ or $\Sigma 29$. The zinc penetration depths for the asymmetrical boundaries are shown in Fig.3, and these show a marked difference from the results on symmetrical boundaries in that $\Sigma 5$ exhibits a minimum penetration depth here, whereas it was the maximum for the symmetrical cases.

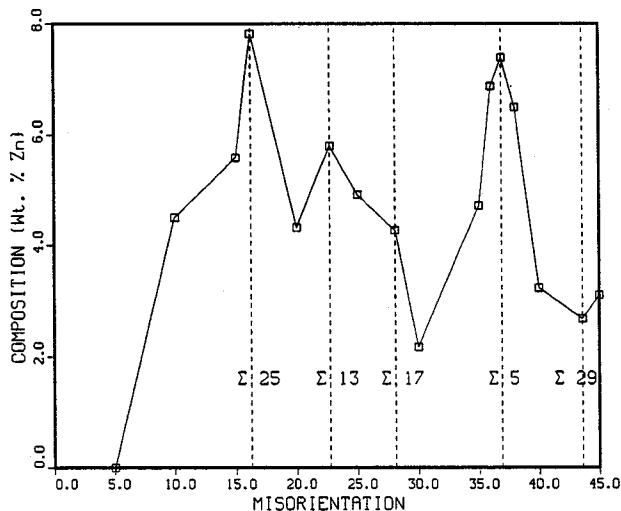


Figure 2. The average zinc concentration deposited by the moving grain boundary, as a function of misorientation, for (100) asymmetrical tilt grain boundaries.

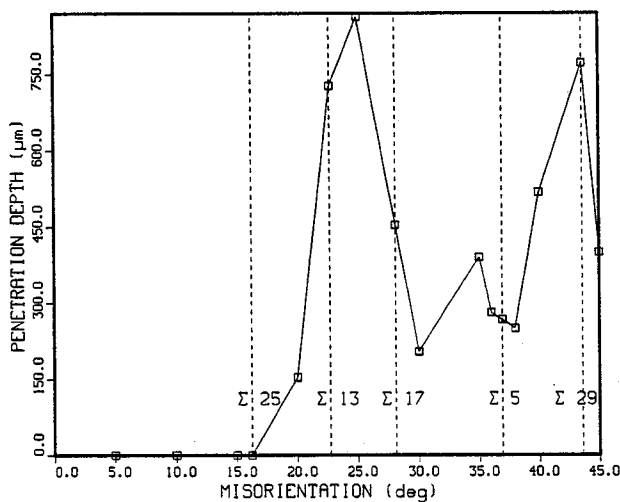


Figure 3. The zinc penetration depth as a function of misorientation for (100) asymmetrical grain boundaries. The minimum for $\Sigma 5$ is in marked contrast with earlier results on symmetrical boundaries.

ii) - (110) Boundaries.

The average migration distances for the (110) asymmetrical grain boundaries are shown in Fig.4. It is interesting to note that in this case a reversal of the migration direction is predicted at approximately 50° of misorientation, at which point the driving force for the coherency strain model becomes zero. In every case the migration direction is as expected on the basis of coherency strain. The magnitudes of the migration distances, however, bear little or no correlation with the theoretical migration rate curve. It is particularly noticeable that the maximum migration distances were obtained for boundaries close to $\Sigma 11$ (50.41°) in spite of the very low driving force at this misorientation, and that small or zero migration distances were measured for boundaries close to $\Sigma 3$ (the primary twin) in spite of large driving forces. In this case, both the $\Sigma 11$ and $\Sigma 3$ boundaries exhibited minimum values of the average solute concentration, but $\Sigma 11$ had a large solute penetration while that in the $\Sigma 3$ boundaries, and those within 10° of that misorientation, had immeasurably small penetration depths.

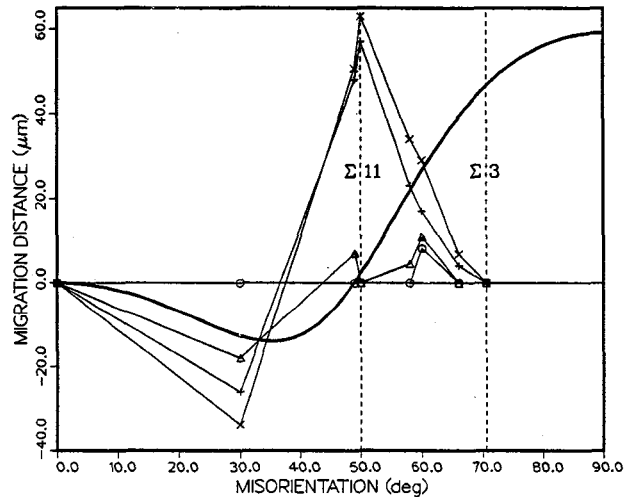


Figure 4. The variation of migration distance with misorientation for (110) asymmetrical tilt boundaries. The four experimental curves correspond to annealing times of 3, 6, 12 and 24 hours. The heavy, continuous curve represents the theoretical variation based upon the coherency strain driving force, and it is fitted to an arbitrary maximum. It is notable that the migration direction is always correctly predicted by the theory.

5 - DISCUSSION

The most striking features of our new results are, first, that the direction of migration is always as predicted by the coherency strain model and, second, that there is excellent correspondence between the predicted migration rates and the measured migration distances for small-to-moderate angle (100) boundaries. These two features add considerably to confidence in the coherency strain mechanism. It is curious, however, to note some interesting comparisons with our earlier work on symmetrical grain boundaries. Among the symmetrical boundaries, small angle grain boundaries were found not to migrate, whereas they do here. It is also interesting that boundaries with well defined structures should so closely obey the predictions of a model that specifically ignores all grain boundary structure. It is also not clear what effect the variation of grain boundary diffusivity with changing misorientation might have, and that is also ignored in the theoretical development. Ultimately, however, the match of the theory with the experiment is compelling, and we conclude that the grain boundary structure has little effect upon DIGM in the small angle regime. This is also readily rationalized in terms of the dislocation climb model, since the lattice dislocations that make up small angle grain boundaries embody no steps [10], so they do not contribute to the migration of the grain boundary. What we observe for the asymmetrical small angle boundaries is therefore "pure" coherency strain and it fits the theory excellently. For the symmetrical small angle boundaries there is also no contribution from coherency strain, because the symmetry ensures that $Y(\mathbf{n}_1) = Y(\mathbf{n}_2)$, so $[Y(\mathbf{n}_1) - Y(\mathbf{n}_2)]$ is zero and no migration is expected. Thus with no coherency strain to drive the migration, and no perturbation from dislocation climb, the symmetrical small angle grain boundaries do not respond to DIGM.

The large angle grain boundaries present other complications. In the symmetrical cases reported earlier, migration should not occur according to the coherency strain model for the reason given above, but in fact it is observed. We attribute this to the nucleation of DIGM via the dislocation climb mechanism for these cases. Once DIGM is nucleated, of course, there is a strain asymmetry across the boundary and it is likely that the grain boundary is no longer in a symmetrical orientation, so the driving force deriving from coherency strain becomes available. It is interesting, however, that DIGM was found to cease for certain well defined asymmetrical boundary planes in the earlier studies [4]. These were found to be planes upon which certain of the available grain boundary dislocations embodied no steps, so the planes were ones on which the dislocation climb mechanism is unavailable. In the present study, the migration rates of large angle grain boundaries showed marked deviations from the coherency strain calculations, and this may be attributed to a variety of causes including lack of diffusivity along the boundary plane, which would prevent DIGM by stopping solute build-up, as discussed below. In addition, of course, there may be contributions to the migration in the large angle regime from the dislocation climb mechanism. This could cause positive or negative deviations from the theoretical curve.

The asymmetrical boundaries studied here present some other new features that were not apparent in the symmetrical boundaries that were studied previously. In particular, in our earlier work [4] we always found that a high solute concentration was associated with a large penetration depth into the grain boundary and a low migration rate, indicating a straightforward competitive relationship between migration of the boundary (and hence deposition of the boundary material) and diffusion along the grain boundary. In the present studies, such a relationship appears not to hold true. For example, the $\Sigma 5$ asymmetrical boundary has a large deposited solute concentration, as it did for the symmetrical cases, but here it shows a large migration distance and a small penetration depth, both of which are contrary to the earlier result on the symmetrical interfaces. Another interesting case is that of the $\Sigma 11$ (110) grain boundary, which shows essentially similar behavior, although the initial grain boundary in this case was considerably faceted along symmetrical grain boundary planes. For both of these boundaries the migration takes an unusual form in the sense that it is not at all continuous. There appears to be a significant incubation time in both cases, extending beyond 6 hours for $\Sigma 11$. Following the incubation period, the boundary moves rapidly to a new position which again appears to be quite stable for long periods of time. It is possible that in these cases the initial grain boundary structure is such as to provide an immobile interface. With the increase in solute concentration in the grain boundary, however, the structure may transform to one that embodies a higher mobility and/or a higher diffusivity. Such transformations were speculated upon by Smith and King [7], and evidence for them has been presented by Sickafus and Sass [11]. Interestingly, $\Sigma 11$ boundaries in f.c.c. metals may have extremely large values of h/b [8] for primitive DSC dislocations. It was speculated [10] and later confirmed experimentally [12] that the stable dislocations in such boundaries would have doubly-primitive DSC Burgers vectors in order to eliminate the energetically expensive steps. In the present study it may be the case that dissociation into primitive DSC dislocations occurs when a certain zinc concentration is attained. If so, the dislocation climb mechanism would promote rapid boundary migration, as observed, for a case with a very small driving force.

The total lack of response of the $\Sigma 3$ -related boundaries appears to relate simply to the low diffusivity in these grain boundaries, which have a core structure that is essentially as close packed as that of the crystal lattice. The energy of the interface is also extremely low, mitigating against the likelihood of a structural transformation such as we postulate for the $\Sigma 5$ and $\Sigma 11$ cases. If the core structure of the $\Sigma 3$ boundary behaves essentially as perfect crystal with respect to diffusion, then as the misorientation changes from the exact coincidence value, the diffusivity should increase just as that of small angle grain boundaries does, because of the increasing density of diffusive "pipes". We see in Fig.4, that as the misorientation decreases from 70.53° , the migration rate of the boundary increases, in spite of a decreasing driving force, according to the coherency strain model. In addition to the increasing diffusivity of the boundary, the boundary mobility may also be increasing with increasing deviation from the coincidence misorientation, as the dislocations that accommodate the orientational deviation also embody steps and may contribute to migration through the dislocation climb mechanism.

6 – SUMMARIZING COMMENTS

It would appear that neither the dislocation climb model nor the coherency strain model can alone explain all of the features of our results. Taken together, however, and understanding the coherency strain as providing the *driving force*, and dislocation climb providing a migration *mechanism*, all of the features of the grain boundary structure dependence can be rationalized. Dislocation climb is the exclusive migration mechanism only for certain interfaces, where migration can be made to cease if it becomes unavailable, but for most grain boundaries more "civilian" forms of atomic transport across the interface can also operate, so we observe the effects of the changing driving force, unconvoluted by changing grain boundary mobility.

ACKNOWLEDGEMENT

This work is supported by the National Science Foundation, under grant number DMR-8901994.

REFERENCES

1. A.H. King: *Int. Mater. Revs.* 32 (1987) 173
2. Y.J. Baik and D.N. Yoon: *Acta Met.* 34 (1986) 2039.
3. W.H. Rhee, Y.D. Song and D.N. Yoon: *Acta Met.* 35 (1987) 57.
4. F.S. Chen and A.H. King: *Acta Met.* 36 (1988) 2827.
5. A.H. King: *Scripta Met.* 21 (1987) 541.
6. C.A. Handwerker, J.W. Cahn, D.N. Yoon, and J.E. Blendell: in *Atomic Transport in Alloys: Recent Developments*, (Ed. G.E. Murch and M.A. Dayananda); 1985, Warrendale, PA, Metallurgical Society of AIME.
7. D.A. Smith and A.H. King: *Phil. Mag.* A44 (1981) 333.
8. A.H. King: *Acta Met.* 30 (1982) 419.
9. F.S. Chen and A.H. King: *Scripta Met.* 21 (1987) 649.
10. A.H. King and D.A. Smith: *Acta Crystall.* A36 (1980) 335.
11. K.E. Sickafus and S.L. Sass: *Acta Met.* 35 (1987) 69.
12. H. Ichinose and Y. Ishida: *Phil. Mag.* A43 (1981) 1253.