



The Effect of Triple Junctions on Grain Boundary Motion and Grain Microstructure Evolution

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Abstract. The theory of steady state motion of grain boundary systems with triple junctions and the main features of such systems are considered. A special technique of in-situ observations and recording of triple junction motion is introduced, and the results of experimental measurements on Zn tricrystals are discussed. It is shown, in particular, that the described method makes it possible to measure the triple junction mobility. It was found that the measured shape of a moving half-loop with a triple junction agrees with theoretical predictions. A transition from triple junction kinetics to grain boundary kinetics was observed. This means that triple junctions can drag boundary motion. It is demonstrated that the microstructural (granular) evolution is slowed down by triple junction drag for any n -sided grain. The second consequence pertains to six-sided grains. For a boundary system with dragging triple junctions there is no unique dividing line between vanishing and growing grains with respect to their topological class anymore, like $n = 6$ in the Von Neumann-Mullins relation.

Keywords: grain boundary, mobility, triple junction, Von Neumann-Mullins relation

Introduction

This paper is dedicated to the grain boundary triple junction mobility, to the problem of triple junction drag on grain boundary motion, or, more correctly, to the problem how grain boundaries and triple junctions move together. We define a “grain boundary triple junction” as a line or a column of intersection of three boundaries. Most frequently such a line of intersection can be met in polycrystals of crystalline solids. The understanding of the fact that such a line (or column) is part of a system with specific thermodynamic properties was realized more than a hundred years ago [1]. However, the kinetic properties of this subject, in

particular, its mobility, were essentially ignored. Although the number of triple junctions in polycrystals is comparable in the order of magnitude with the number of boundaries, all peculiarities in behaviour of polycrystals during grain growth have been traditionally referred to grain boundary motion. Actually, the mean grain size, which is usually defined as the mean spacing of grain boundaries can be equally well associated with the mean distance between the triple junctions.

However, until recently it was tacitly assumed both in theoretical approaches, computer simulations and interpretation of experimental results that triple junctions do not disturb grain boundary migration and that their role in grain growth is reduced to preserve

the thermodynamically prescribed equilibrium angles where boundaries meet. At the same time it was shown [2] that the motion of triple junctions under the driving force caused by the grain boundary surface tension might involve an additional dissipation of energy, in other words, triple junctions may have a finite mobility. The main reasons for our poor knowledge on triple junctions are the difficulties to conduct experiments in the course of which the grain boundaries and triple junction(s) move in steady state. The crucial point is that the stationary motion of a grain boundary system with a triple junction is possible only in a very narrow class of geometrical configurations [2, 3].

Firstly, a phenomenological approach will be considered. We will cover the configurations in which a steady state motion of a grain boundary system with a triple junction is feasible. In particular, it will be analyzed how the angles at the tip of a triple junction depend on the mobilities of grain boundaries, triple junctions and grain size. Furthermore, experimental evidence of the finite mobility of a triple junction and its drag effect will be presented. Finally, we will show that the existence of triple junctions with a finite mobility dramatically affects the kinetics of grain growth and that the interaction of moving grain boundaries with triple junctions drastically changes our conception of microstructure evolution and, in particular, the theoretical basis of grain growth in 2D systems—the Von Neumann-Mullins relation.

**Triple Junction Motion
(Phenomenological Approach)**

In references [2, 3] model 2D grain boundary systems were considered (Figs. 1 and 2). As can be seen from Fig. 1, such model configuration correlates with grains in a polycrystal with less than 6 neighbors (adjacent grains), in other words, the topological class of the grain is smaller than 6. The main assumptions of this consideration are: (i) all grain boundaries have equal mobilities and surface tensions, irrespective of their misorientation and crystallographic orientation of the boundary in the crystal; (ii) the mobility of a grain boundary is independent of its velocity; (iii) the normal grain boundary displacement rate v is equal to

$$v = \sigma m_b K \equiv A_b K \tag{1}$$

where m_b is the grain boundary mobility, σ is the grain boundary surface tension, K is the local curvature of

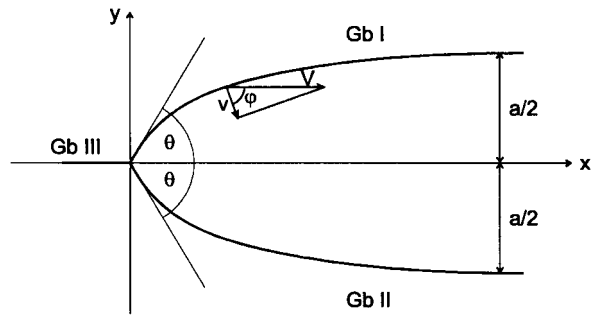


Figure 1. Configuration of grain boundaries at a triple junction during steady state motion for $n < 6$.

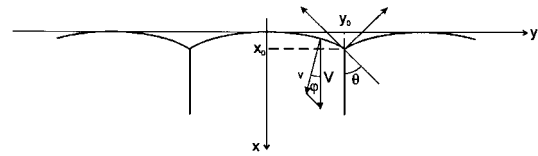


Figure 2. Configuration of grain boundaries at triple junctions during steady state motion for $n > 6$.

the grain boundary

$$K = \frac{d\varphi}{d\ell} = -\frac{y''}{[1 + (y')^2]^{3/2}} \tag{2}$$

where φ is tangential angle at any given point of the grain boundary, $d\ell$ is an element of the grain boundary perimeter and $y(x)$ is the shape of the boundary. During steady state motion of the whole system the velocity V parallel to the x -axis (Figs. 1 and 2) is related to the rate of normal displacement v :

$$v = V \cos \varphi = V \frac{y'}{[1 + (y')^2]^{1/2}} \tag{3}$$

where $y(x)$ is the shape of the positive part (upper part in Fig. 1) of the curved boundary. (Due to the mirror symmetry of the problem relative to the x -axis, the shape of the lower (left in Fig. 2) boundary is the negative equivalent.)

From Eqs. (1)–(3) we obtain the equation for the steady-state shape of the moving grain boundary

$$y'' = -\frac{V}{m_b \sigma} y' [1 + (y')^2] \tag{4}$$

Three boundary conditions permit us to find the shape $y(x)$ and velocity V of the moving grain

boundary (Fig. 1)

$$y(0) = 0 \quad (5)$$

$$y(\infty) = \frac{a}{2} \quad (6)$$

$$y'(0) = \tan \theta \quad (7)$$

The meaning of the length a and the angle θ is clear from Fig. 1. A driving force $\sigma(2 \cos \theta - 1)$ acts on the triple junction from the curved boundaries. Introducing the mobility of the triple junction m_{ij} , its velocity reads

$$V = m_{ij}\sigma(2 \cos \theta - 1) \quad (8)$$

Due to the fact that the driving force acting on the grain boundary is a pressure and the driving force on the triple junction is a force, the dimensions of grain boundary and triple junction mobility are different, so their ratio m_b/m_{ij} has the dimension of a length.

For configuration in Fig. 1 Eqs. (4)–(8) define the problem completely. The solution can be expressed as

$$y(x) = \xi \arccos(e^{-x/\xi+c_1}) + c_2 \quad (9)$$

with

$$\xi = \frac{a}{2\theta}$$

$$c_1 = \frac{1}{2} \ln(\sin \theta)^2$$

$$c_2 = \xi \left(\frac{\pi}{2} - \theta \right)$$

The velocity V of steady state motion of the system is

$$V = \frac{2\theta m_b \sigma}{a} \quad (10)$$

The steady state value for the angle θ can be found from the equation

$$\frac{2\theta}{2 \cos \theta - 1} = \frac{m_{ij} a}{m_b} = \Lambda \quad (11)$$

If a triple junction is mobile and does not drag grain boundary motion the dimensionless criterion $\Lambda \rightarrow \infty$ and $\theta \rightarrow \pi/3$, i.e. the equilibrium angular value at a triple junction in the uniform grain boundary model. In contrast, however, when the mobility of the triple junction is relatively low (strictly speaking, when $m_{ij} \ll m_b$) then $\theta \rightarrow 0$ (Fig. 3). It should be stressed that the angle θ is strictly defined by the dimensionless criterion Λ , which, in turn, is a function of not only the ratio

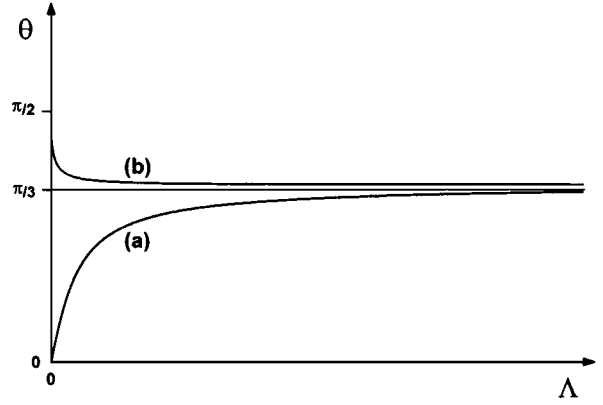


Figure 3. Angle θ as a function of Λ . (a) for $n < 6$, Eq. (11); (b) for $n > 6$, Eq. (17).

of triple junction and grain boundary mobilities, but on the grain size as well.

The steady state motion of a grain boundary system with a triple junction shown in Fig. 2 is determined by the system of Eqs. (1)–(4) only with different boundary and initial conditions [3]

$$y'(0) = \infty$$

$$y'(x_0) = \tan \theta \quad (12)$$

$$y(0) = 0$$

The velocity of the triple junction motion can be read as (Fig. 2)

$$V_{ij} = m_{ij}\sigma(1 - \cos \theta) \quad (13)$$

Like in the previous case the Eqs. (4), (12), (13) define the problem completely

$$y(x) = -\frac{x_0}{\ln \sin \theta} \arccos(e^{(x/x_0) \ln \sin \theta}) \quad (14)$$

The velocity of steady state motion of the system is

$$V = \frac{m_b \sigma}{x_0} \ln \sin \theta \quad (15)$$

The length x_0 replaces the role of the grain size a in the previous case (Fig. 2) or

$$\begin{aligned} y_0 = y(x_0) &= -\frac{x_0}{\ln \sin \theta} \arccos(e^{\ln \sin \theta}) \\ &= -\frac{x_0}{\ln \sin \theta} (\pi/2 - \theta) \end{aligned} \quad (16)$$

The dimensionless criterion Λ for the considered configuration can be obtained from Eqs. (13), (15) and describes as in the previous case the influence of the triple junction mobility on grain boundary motion

$$-\frac{\ln \sin \theta}{1 - 2 \cos \theta} = \frac{m_{\text{tj}} x_0}{m_b} = \Lambda \quad (17)$$

One can see that for $\Lambda \gg 1$, when the boundary mobility determines the kinetics of the system ($m_{\text{tj}} \gg m_b$) the angle θ tends to its equilibrium value ($\pi/3$).

Again, the angle θ changes when a low mobility of the triple junction starts to drag the motion of the boundary system. However, as can be seen from Eq. (17) and Fig. 3, in this case the steady state value of the angle θ *increases* as compared to the equilibrium state. For $\Lambda \ll 1$ the angle θ tends to approach $\pi/2$ (Eq. (17), Fig. 3).

Triple Junction Motion: Experimental Results

As can be seen from Figs. 1 and 2 and Eqs. (8)–(11) and (13)–(17) the consideration can be applied not only to the uniform grain boundary model, but to the so-called symmetrical configuration as well, in which the curved grain boundaries are the same and different from the straight boundaries. This approach represents the theoretical background of a measurements of the velocity of motion of a grain boundary system with a triple junction and of the triple junction mobility [4].

In the following a boundary system as shown in Fig. 1 with two identical curved boundaries (GB I and II) and a different straight boundary (GB III) will be considered.

The respective surface tensions and mobilities of the boundaries are

$$\begin{aligned} \sigma_1 &= \sigma_2 \equiv \sigma \neq \sigma_3 \\ m_{b1} &= m_{b2} \equiv m_b \neq m_{b3} \end{aligned} \quad (18)$$

The velocity of the triple junction V_{tj} can be expressed as [5]

$$V_{\text{tj}} = m_{\text{tj}} \sum \sigma_i \tau_i \quad (19)$$

where τ_i is the unit vector normal to the triple line and aligned with the plane of the respective boundary i . If the angles at the triple junction are in equilibrium, the driving force is equal to zero, and for a finite triple junction mobility the velocity V_{tj} should vanish as well. Consequently, for a finite m_{tj} the motion of the triple

junction disturbs the equilibrium of the boundaries. For the situation given in Fig. 1

$$V_{\text{tj}} = m_{\text{tj}} (2\sigma \cos \theta - \sigma_3) \quad (20)$$

From Eqs. (20) and (10) we arrive at the relation for the criterion Λ for the case of a symmetrical configuration

$$\frac{2\theta}{2 \cos \theta - \sigma_3/\sigma} = \frac{m_{\text{tj}} a}{m_b} = \Lambda \quad (21)$$

The criterion Λ , as mentioned before, reflects the drag influence of the triple junction on the migration of the system. For a low mobility of the triple junction ($\Lambda \rightarrow 0$) the motion of the system is controlled by the mobility of the triple junction, and θ tends to zero. For the opposite limiting situation ($\Lambda \rightarrow \infty$) the motion of the system is independent of the triple junction mobility and is governed by the grain boundary mobility. The velocity of the motion of the boundary system is then given by

$$V = \frac{2\theta_{\text{eq}} m_b \sigma}{a} \quad (22)$$

where the equilibrium triple junction angle θ_{eq} is equal to

$$\theta_{\text{eq}} = \arccos\left(\frac{\sigma_3}{2\sigma}\right) \quad (23)$$

The two states of motion of the entire grain boundary system can be distinguished experimentally for a given ratio σ_3/σ by measuring the contact angle θ .

The experiments were carried out on zinc-tricrystals with a grain boundary geometry as shown in Fig. 1 [4]. The tricrystals were produced of high purity Zn (99.999 at.%) by a technique of directional crystallisation (Fig. 4). The orientations of the three adjacent grains of each sample were determined by the Laue-technique, electron back scatter diffraction (EBSD) and by an investigation of the fracture surface of a cracked sample. For the latter method small cracks were induced by a sharp knife in the sample cooled to liquid nitrogen temperature. The cracks propagated along the basal plane of each grain. Hence, the misorientation could be determined by the orientation of the surface of the cracks.

For measuring the migration rate and the geometry of the grain boundary system during the motion at elevated temperatures a modified optical microscope operating with polarised light and a hot stage with a

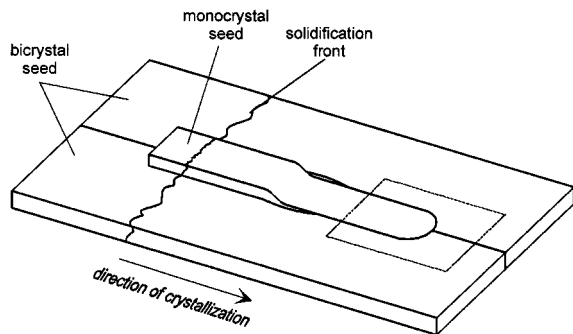


Figure 4. Method of tricrystal fabrication; the rectangular area represents the tricrystal used for migration experiments.

protecting nitrogen gas atmosphere was used. An additional polarisation filter applied in the reflected beam allowed to distinguish the different orientations of the grains by the different intensity of the reflected light. A colour video camera was attached to the microscope and connected to a video cassette recorder to record the motion of the triple junction system during the experiment. For each temperature the velocity of the triple junction system, the angle θ and the width a of the vanishing grain were measured. For this, single video frames were grabbed by a computer and in accordance with Eq. (9) a computed shape of the grain boundary system was superimposed. By fitting the computed shape to the observed shape, the angle θ and the width a of the shrinking grain were determined.

The motion of different triple junctions with a grain boundary configuration as in Fig. 1 were investigated in the temperature range between 330 and 405°C [4]. The triple junctions of one set were formed by two nearly identical high angle (01 $\bar{1}$ 0) tilt grain boundaries (curved boundaries, GB I and II) and a low angle tilt boundary (straight boundary, GB III). Due to the different properties of low angle grain boundaries and high angle boundaries, the triple junctions of this set of the samples were symmetrical junctions as described above. Under the assumption that the properties of a high angle boundary vary only little with misorientations, the behaviour of the triple junctions of the second set of samples can be modelled as an ideal triple junction according to [2]. For both types of triple junctions the shape of the moving grain boundary system was similar to the shape predicted by theory. Figure 5 shows a series of video frames of a moving symmetrical triple junction. The straight grain boundary (GB III) is invisible due to the small orientation difference (3°) of the adjacent grains. The solid line in the lower right picture was computed in accordance with Eq. (9) and

fitted the shape of the curved grain boundaries quite well [4]. For so-called “ideal” triple junctions the same behaviour was observed [4].

For all samples the velocities V (Fig. 6) and the angles θ (Fig. 7) were found to be constant for a given temperature over the entire investigated temperature range. Evidently, the assumption of a steady state motion of the entire grain boundary system was justified.

The angle θ increased with increasing temperature. In particular for the symmetrical triple junction the change of θ was drastic (Fig. 8). In accordance with the temperature dependence of θ , the criterion Λ , determined by Eqs. (11) and (21), was found to be constant for a given temperature, but increased with increasing temperature (Figs. 9 and 10). At low temperatures Λ was on the order of unity and increased with rising temperature up to 3 orders of magnitude. For the calculation of Λ for symmetrical triple junctions the ratio σ_3/σ was determined under the assumption that for temperatures near the melting point the value of θ reaches the thermodynamic equilibrium value (Eq. (23)).

For the first time two different regimes of coupled triple junction-grain boundary motion were observed, indicated by a change of the angle θ with temperature. At low temperatures, where Λ is on the order of unity (Figs. 9 and 10), the motion of the boundaries is dragged by the hardly mobile triple junction. Accordingly, the angle θ is smaller than predicted by the equilibrium of grain boundary surface tensions (Figs. 5 and 8), and the motion of the entire boundary system is controlled by the mobility of the triple junction. With increasing temperature the triple junction becomes more mobile compared to the grain boundary mobility as indicated by an increasing value of Λ (Figs. 9 and 10). Therefore, the drag of the triple junction decreases and at high temperatures, close to the melting point, the motion of the entire boundary system is governed by the grain boundary mobility of the curved boundaries only. As a consequence of the transition of the state of motion the angle θ tends to attain its thermodynamic equilibrium value with increasing temperature (Fig. 7). As shown in [4], such temperature dependence of θ can not be explained in terms of a different temperature coefficient of the grain boundary surface tension. Consequently, the temperature dependence of θ for all samples must result from the change of the kinetics of motion, reflected by the temperature dependence of Λ (Figs. 8 and 9).

It is believed that the experiment [6] unambiguously proves the existence of a specific mobility of triple junctions. The dimensionless criterion Λ specifies the

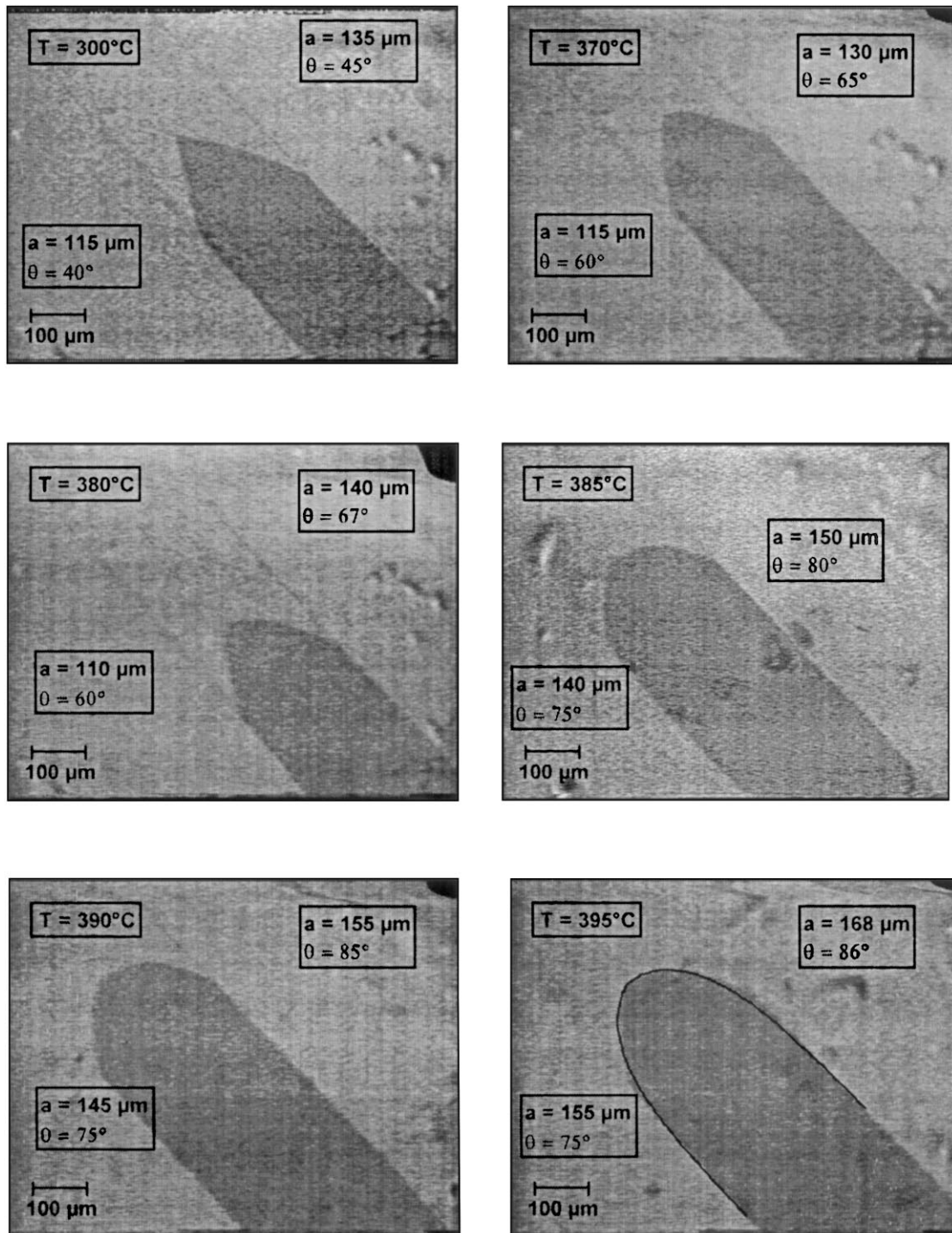


Figure 5. Video frames for different temperatures of a moving symmetrical triple junction. GB III (s. Fig. 1) is invisible, located at the tip of the two visible boundaries. At $T = 300^\circ\text{C}$ the grain boundary system did not move at all. The solid line generated according to Eq. (9) in the lower right frame fits the boundary shape.

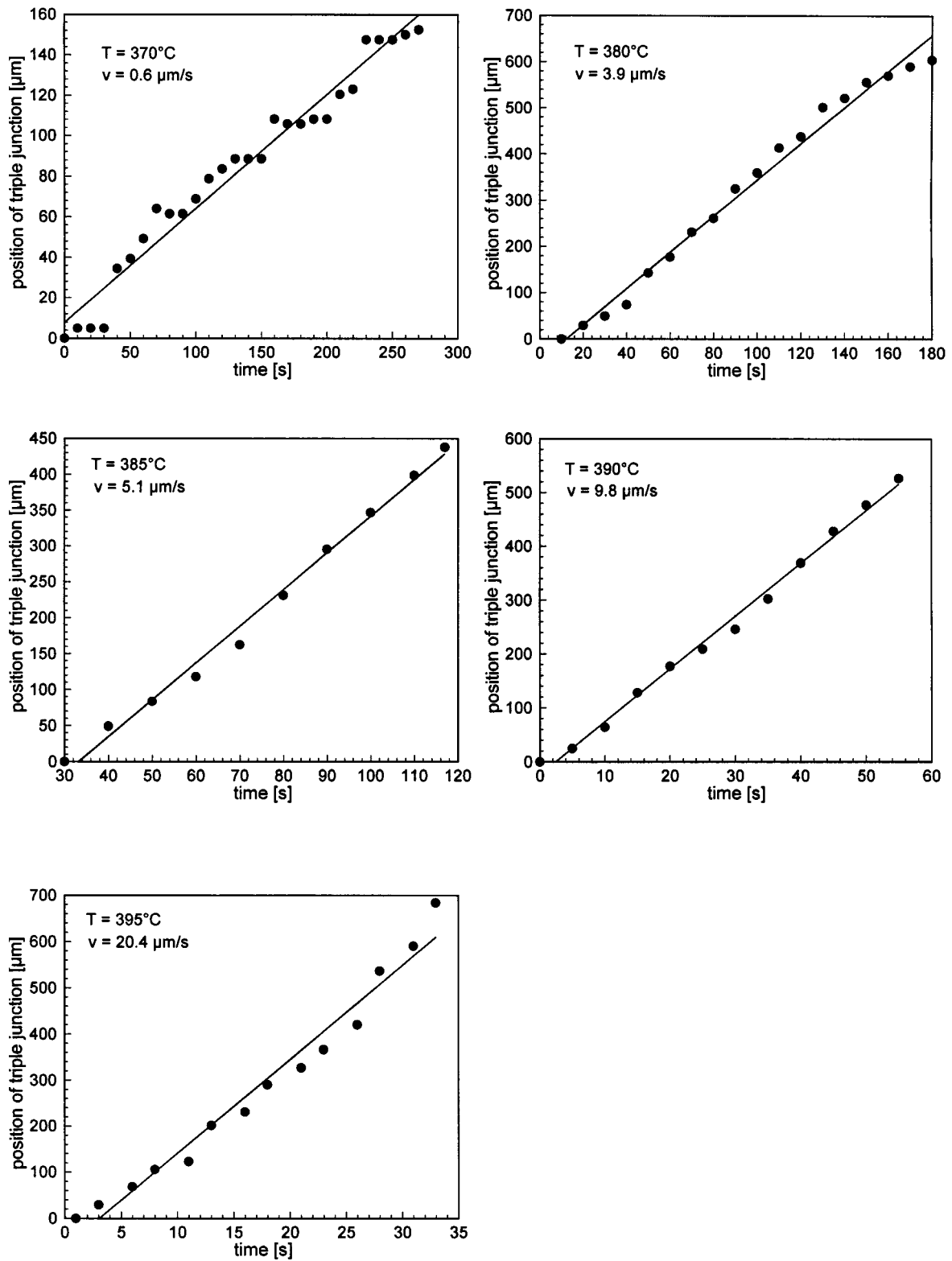


Figure 6. Triple junction position vs. time for different temperatures for a symmetrical triple junction.

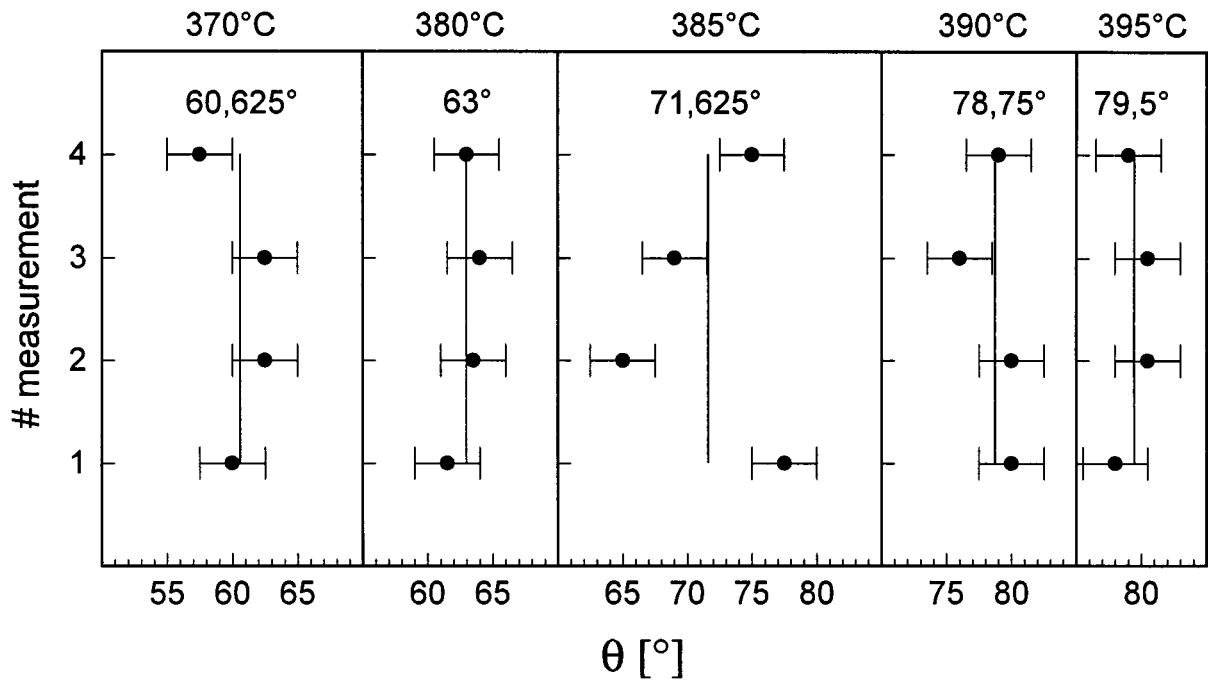


Figure 7. Reproducibility of measurement of the angle θ at different temperatures.

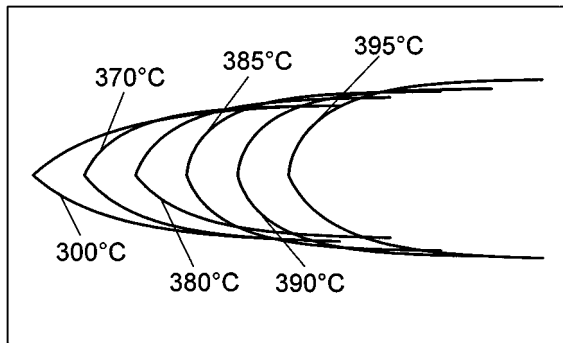


Figure 8. Evolution of the shape of the grain boundary system of a sample with symmetrical triple junction with increasing temperature.

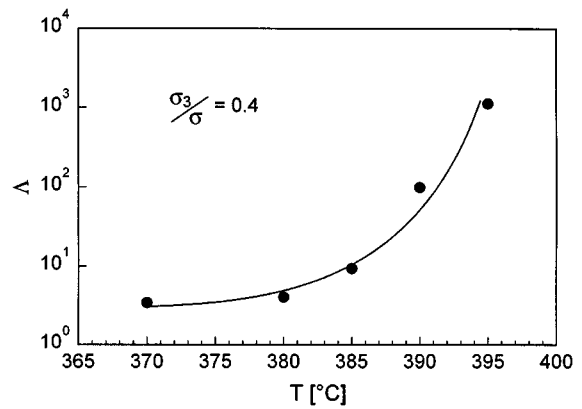


Figure 9. Temperature dependence of the criterion Λ for a symmetrical triple junction.

ratio of triple junction mobility to grain boundary mobility (Eq. (11)). For low temperatures Λ is on the order of unity and thus, the mobility of triple junctions is comparable to the grain boundary mobility (normalized by the width a of the shrinking grain).

For all investigated triple junctions the activation enthalpy of triple junction migration H_{tj} was found to be higher than for grain boundary migration H_b . This difference is caused by the observed temperature dependence of Λ . As mentioned above at low

temperatures the motion of the boundary system is governed by the reduced mobility of the triple junction. With increasing temperature the triple junction becomes more mobile compared to the grain boundary due to the higher activation enthalpy of triple junction migration. At high temperatures the motion of the boundary system is governed by the process with the lower activation enthalpy, in this case the boundary mobility. But even at high temperatures the motion of the boundaries is not totally free. A triple junction with

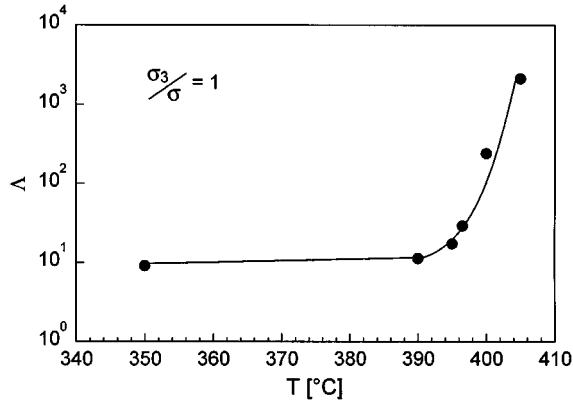


Figure 10. Temperature dependence of the criterion Δ for an ideal triple junction.

finite mobility, as revealed by the presented results, can only move under the action of a driving force. The driving force for the motion of a triple junction results from the imbalance of the angle θ (Eq. (10)) and reduces the driving force for grain boundary motion (Eq. (8)). Hence, the motion of boundaries in a system like in the current investigation is always dragged by the triple junction [4].

The Effect of Triple Junction Drag on Grain Growth

The Von Neumann-Mullins relation [7, 8] forms the basis for practically all theoretical and experimental investigations as well as computer simulations of microstructure evolution in 2D polycrystals in the course of grain growth [9–11].

One of the principal features underlying this relation is an assumption that the triple junctions do not drag grain boundary motion. However, the experimental results considered above contradict this condition.

The fundamental Von Neumann-Mullins relation is based on three essential assumptions, namely a uniform grain boundary, independence of grain boundary mobility of its velocity and no effect of triple junctions on grain boundary motion; therefore, the angles at triple junctions are in equilibrium and, within the uniform grain boundary model, equal to 120° .

Let us consider a 2D grain with an area S (Fig. 11) [5]. In the time interval dt all points on the grain boundaries of the considered grain will displace normal to the grain boundaries by the amount Vdt , where V is the grain boundary migration rate. Accordingly, the rate

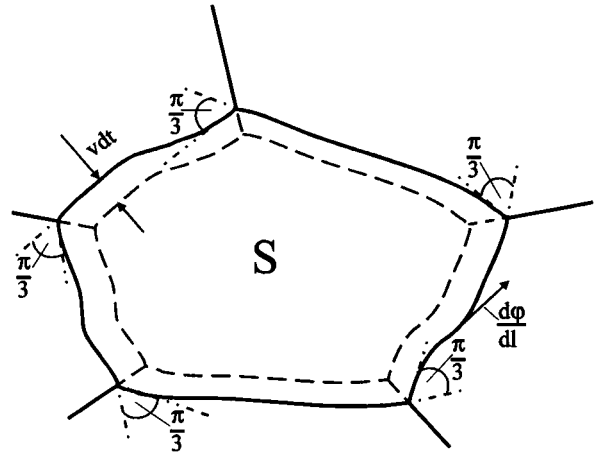


Figure 11. Definition of parameters for the effect of triple junctions for a calculation of the rate of grain area change.

of change of the grain area S can be expressed by

$$\frac{dS}{dt} = \oint v dl \quad (24)$$

where dl is an element of the grain perimeter. For grain growth the normal grain boundary velocity satisfies Eqs. (1) and (2).

From from Eqs. (1), (2), and (25) follows

$$\frac{dS}{dt} = -A_b \oint d\varphi \quad (25)$$

If the grain were bordered by a smooth line, the integral in Eq. (25) would equal 2π . However, owing to the discontinuous angular change at every triple junction, the angular interval $\Delta\varphi = \pi/3$ is subtracted from the total value 2π for each triple junction. Consequently

$$\frac{dS}{dt} = -A_b \left(2\pi - \frac{n\pi}{3} \right) = \frac{A_b\pi}{3} (n - 6) \quad (26)$$

where n is the number of triple junctions for each respective grain, i.e. the topological class of the grain. Thus, the rate of area change is independent of the shape of the boundaries and determined by the topological class n only. Grains with $n > 6$ will grow and those with $n < 6$ will disappear [8].

Let us consider how the mentioned results influence the Von Neumann-Mullins relation (26). For this we assume that the influence of the triple junctions is rather large, but, nevertheless, the motion of the system can

be viewed as grain boundary motion, since the driving force is still due to the grain boundary curvature, i.e. the role of the triple junctions is reduced to a change of the angle θ . As mentioned above Eq. (11) describes the steady state value of the angle θ . Of course, triple junctions in real polycrystals rarely experience steady state motion. However, the attainment of a true steady state is not important in this context. Even if the angle θ is not in steady state with the moving triple junction, it will be different from the equilibrium angle $\theta = \pi/3$ as assumed for the Von Neumann-Mullins relation and thus, affect the kinetics with the same tendency as in steady state.

The rate of area change for a grain with $n < 6$ (Fig. 1) can be expressed as:

$$\begin{aligned} \frac{dS}{dt} &= -m_b \sigma \oint d\varphi = -A_b [2\pi - n(\pi - 2\theta)] \\ &= A_b (\pi - 2\theta) \left(n - \frac{2\pi}{\pi - 2\theta} \right) \end{aligned} \quad (27)$$

Since the limited mobility of the triple junction reduces the steady state value of the angle θ as compared to the equilibrium angle, the shrinking rate of grains with $n < 6$ decreases, as obvious for the case when the mobility of the triple junction becomes very low. In other words, for grains with $n < 6$ the influence of the triple junction mobility slows down the process of grain structure evolution, decreasing the vanishing rate of grains with small topological class ($n < 6$).

For grains with topological class greater than 6 let's refer to the considered steady state motion of a grain boundary system with a large number of triple junctions (Fig. 2) [3]. The dimensionless parameter Λ , which describes the influence of the triple junction mobility on grain boundary migration for such a system is given by Eq. (17). When a low mobility of the triple junction starts to drag the motion of the boundary system, the angle θ changes. However, in this case the steady state value of the angle θ *increases* as compared to the equilibrium state. Such an increase of the angle θ also decreases the magnitude of $(\pi - 2\theta)$ in Eq. (27), in other words, it decreases the "effective" magnitude of the topological class of the growing grain with $n > 6$. Consequently, microstructural evolution will slow down due to triple junction drag for any n -sided grain.

The only exception holds for $n = 6$, since a hexagonal grain structure becomes unstable when the contact angle $2\theta \neq 2\pi/3$. Since the actual magnitude of θ is

determined by the triple junction and grain boundary mobility as well as the grain size and is independent of the number of sides of a grain, *there is no unique dividing line between vanishing and growing grains with respect to their topological class anymore, like $n = 6$ in the Von Neumann-Mullins approach.*

Conclusions

Experimental results of triple junction motion in Zn tricrystals are reported. It is shown that triple junctions have a finite mobility and thus, can drag grain boundary motion. The effect of triple junction mobility on the rate of change of the grain area during grain growth was investigated. It was found that a finite junction mobility exerts a drag on the adjoining grain boundaries. This is reflected by a deviation of the grain vertex angles at triple junctions from their equilibrium value $2\pi/3$ and correspondingly, by a modification of the Von Neumann-Mullins relation. It was shown that for the situation when the triple junction influence on grain boundary motion is large enough, but nevertheless the grain boundary motion is controlled by grain boundary kinetics, the triple junction influence results in a reduced rate of microstructure evolution during grain growth. One of the main consequences of the consideration relates to the stability of a hexagonal grain structure under the conditions when the low mobility of the triple junctions drags the grain boundary motion. Since the actual magnitude of the angle at the tip of a triple junction is determined by the triple junction and grain boundary mobility as well as the grain size and does not depend on the number of sides of a grain, there is no unique border line between vanishing and growing grains with respect to their topological class anymore, like $n = 6$ in the Neumann-Mullins approach.

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