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Kinematics of connected grain boundaries in 2D

The motion of connected boundary systems was investigated in Al and Zn tricrystals. It was found that triple junctions can exert a drag force on the adjoining boundaries owing to a lower mobility. This drag manifests itself by a deviation of the dihedral angles at the triple junction from the equilibrium angles due to grain boundary surface tensions. The strength of triple junction drag can be expressed in terms of a criterion $\Lambda = (m_{tj} \cdot a/m_b)$, where m_{tj} is the triple junction mobility, m_b the boundary mobility, and a is the grain size. Therefore, the effect should be particularly pronounced in fine-grained and nanocrystalline materials. It is shown that the von Neumann–Mullins relation of 2D grain growth is modified by triple junction drag such that there is no unique relationship between growth rate and the number of sides anymore. For very small Λ, the triple junctions control grain structure evolution during grain growth, which leads to polyhedral grain shapes with flat boundaries. The theoretical concept is supported by computer simulations of grain growth and in-situ annealing experiments in the SEM.

Keywords: Grain boundaries; Triple junctions; Mobility; Von Neumann-Mullins relation; Grain growth

1. Introduction

The contiguous arrangement of grains in a polycrystal requires that their internal interfaces are connected, i. e., form junctions, for instance, triple junctions in 2-D polycrystals or triple lines, and quadrupel-junctions in 3-D polycrystals. It is conventional conception that these junctions do not play a role for the kinetic behavior of the connected boundary system, but obviously both boundaries and junctions have to move jointly to comply with the conservation of contiguity, i. e., of boundary connectivity. This study is concerned with the mobility of grain boundary triple junctions and its effect on grain structure evolution.

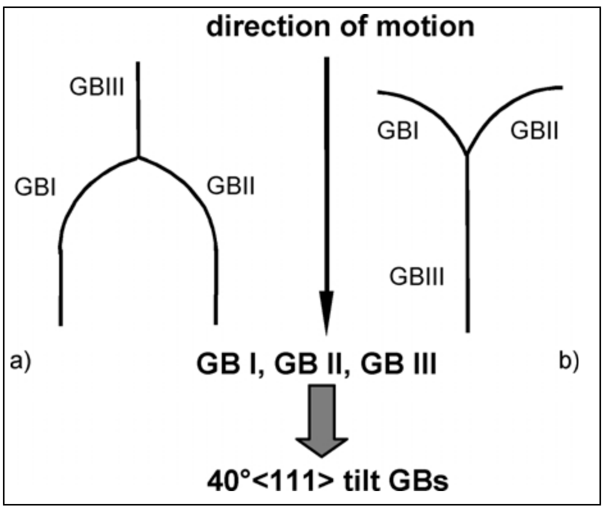

Fig. 1. The two basic configurations of triple junctions with steady state motion.

2. Tricrystal experiments

Individual triple junctions are best realized in tricrystals. Actually, there are only two different configurations that have to be distinguished in 2-D tricrystals (Fig. 1). In one configuration (Fig. 1a), one of the boundaries is extended, like during the shrinkage of small grains, and in the other configuration (Fig. 1b), one grain boundary is eliminated like during the growth of large grains.

Let us consider the kinetic behavior of the first configuration (Fig. 2). The curved boundaries experience a force to move towards their center of curvature which results in a constant horizontal velocity V_b (Fig. 2a), which can be derived from the shape function of the boundaries as

$$V_b = \frac{2\Theta m_b \sigma}{a} \tag{1}$$

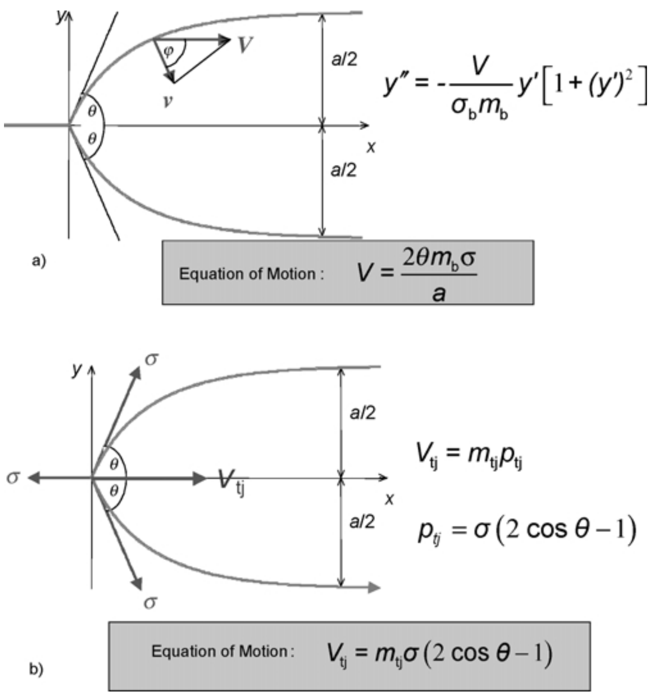

Fig. 2. Equation of motion for connected boundaries; a) boundary motion; b) triple junction motion.

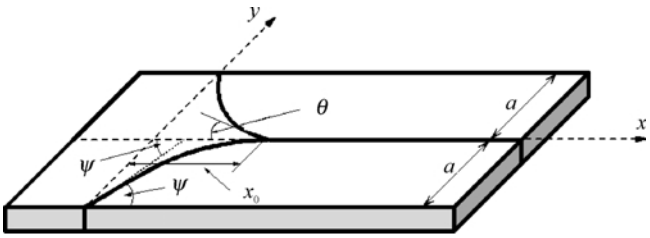

Fig. 3. Tricrystal geometry for configuration (b) in Fig. 1 with retracting boundary.

where Θ is half the dihedral contact angle at the triple junction, m_b the boundary mobility, σ the grain boundary surface tension, and a the thickness of the shrinking grain. On the other hand, the triple junction will experience a force to move if the three grain boundary surface tensions acting on it, do not add up to zero (Fig. 2b), or

$$V_{tj} = m_{tj}\sigma(2\cos\Theta - 1) \qquad (2)$$

Since both boundaries and triple junction have to move jointly, i. e., with the same speed, we obtain from Eq. (1) and Eq. (2)

$$\frac{2\Theta}{2\cos\Theta - 1} = \frac{m_{tj}a}{m_b} \equiv \Lambda \qquad (3)$$

This defines the criterion Λ which reflects essentially the ratio of junction to boundary mobility but also depends on grain size a.

For the second boundary configuration, shown in Fig. 1b, we obtain from the same consideration (Fig. 3)

$$\Lambda = \frac{m_{tj}x_0}{m_b} = \frac{\ln\dfrac{\sin\Theta}{\sin\psi}}{1 - 2\cos\Theta} \qquad (4)$$

where x_0 is a characteristic lateral dimension of the grain (comparable to the grain size), and ψ is the inclination of the boundary at the surface.

For $\Lambda \gg 1$, the angle $\Theta \to 60°$, the junction mobility becomes very high, $m_{tj} \to \infty$, hence, the migration rate is determined by the boundary velocity

$$V = \frac{2\pi\, m_b\sigma}{3a} \qquad (5a)$$

By contrast, if Λ is small ($\Lambda \to 0$), the angle $\Theta \to 0$ and correspondingly $m_{tj} \to 0$. In this case, the junction mobility controls the kinetics of the system which moves with the velocity

$$V = m_{tj}\sigma \qquad (5b)$$

Experiments on Zn [1, 2] and Al [3] have shown that Θ indeed changes with temperature, such that the equilibrium dihedral angle is only attained at sufficiently high temperatures, whereas it is markedly smaller at lower temperatures (Fig. 4), which manifests itself as a strong increase of Λ with increasing temperature (Fig. 5a). An activation analysis of both temperature regimes reveals that the triple junction moves with a higher activation energy (Fig. 5b). Owing to the dependence of Λ on grain size, the change from boundary control to junction control will occur at the higher temperatures the smaller the grain size.

3. Grain growth kinetics

Triple junctions do affect the kinetics of grain growth, which is most adequately described in 2-D by the von Neumann–Mullins equation [4–6] (Fig. 6)

$$\frac{dS}{dt} = \frac{A_b\pi}{3}(n - 6) \qquad (6)$$

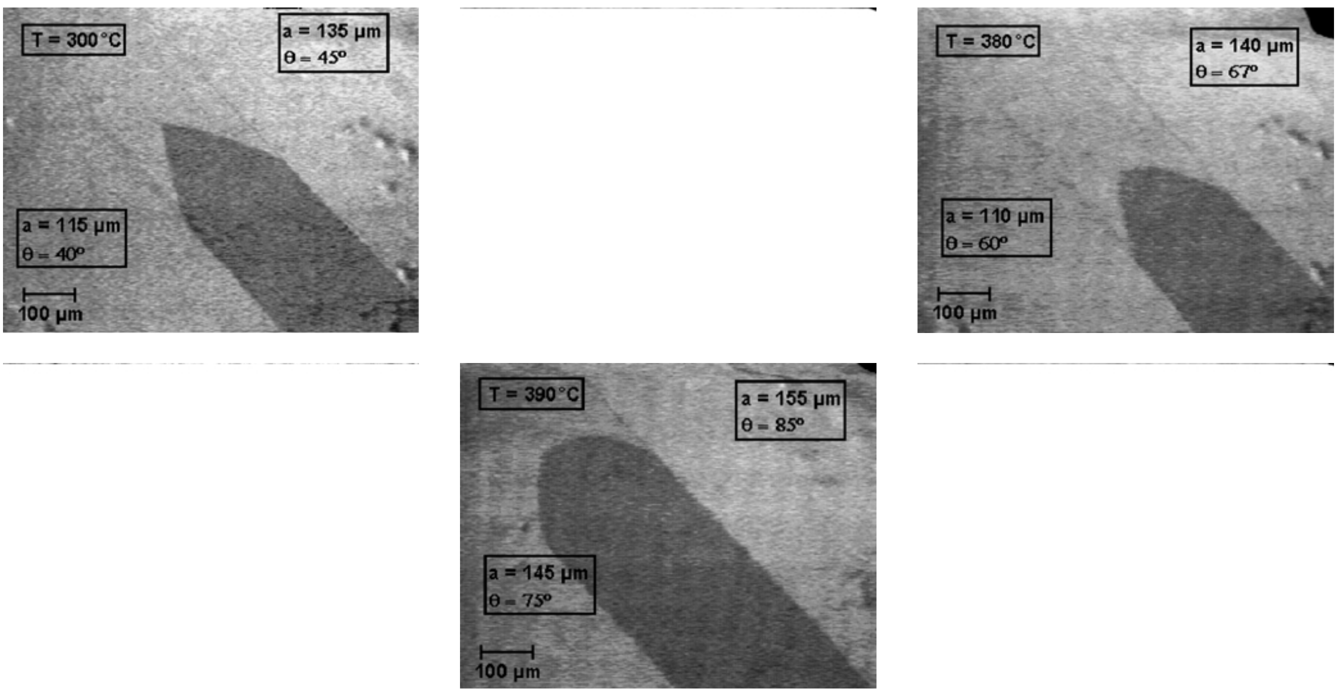

Fig. 4. Optical micrographs of triple junction motion in Zn [1]. The 3rd boundary at the triple junction is a low-angle boundary and, thus, invisible. The dihedral contact angle notably increases with increasing temperature.

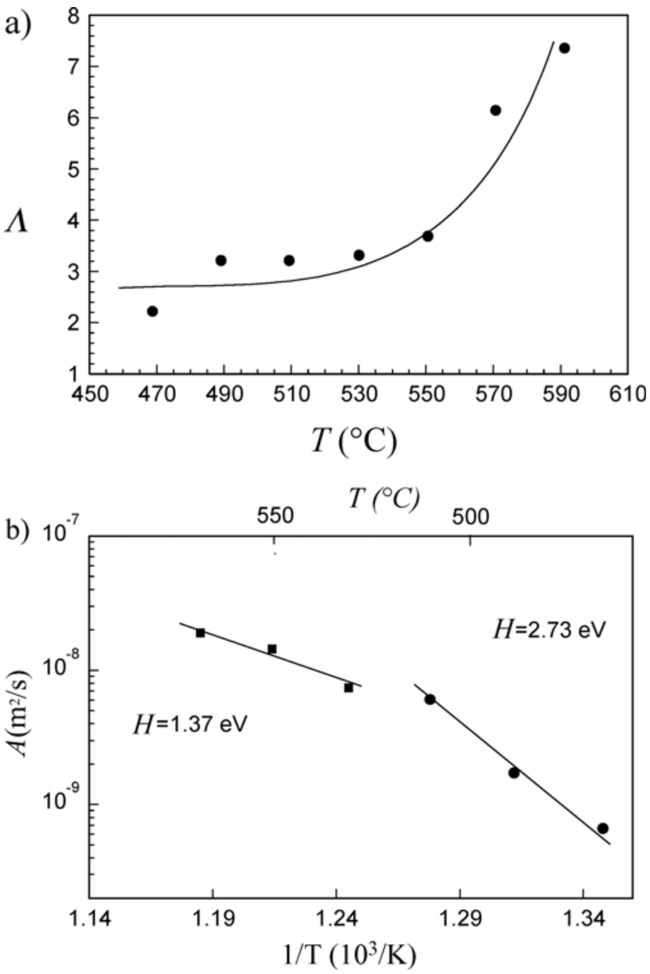

Fig. 5. $A(T)$ and Arrhenius plot of the mobility of a connected boundary system in Al tricrystals.

where S is the area of a considered grain, $A_b = m_b\sigma$ is the reduced mobility, and n is the number of sides of a grain. If the triple junctions exert a drag on the adjoining grain boundaries owing to their lower mobility, it can be derived that for $n < 6$

$$\frac{dS}{dt} = \frac{A_b[2\pi - n(\pi - 2\Theta)]}{1 + \dfrac{2\cos\Theta - 1}{2\Theta}} \tag{7a}$$

and for $n > 6$

$$\frac{dS}{dt} = \frac{A_b[n(\pi - 2\Theta) - 2\pi]}{1 - \dfrac{1 - 2\cos\Theta}{\ln(\sin\Theta)}} \tag{7b}$$

Since Θ is a function of Λ, the growth rate can be also expressed in terms of Λ. Of particular interest is the case $\dot{S} = 0$, which occurs for $n^* = 6$ in the von Neumann–Mullins case. As obvious from Fig. 7, under the action of triple junction drag there is no unique n^* anymore, but n^* depends on Λ, and there are different branches, n_h^* and n_ℓ^* for $n > 6$ and $n < 6$, respectively. For small Λ, there is a large gap between n_ℓ^* and n_h^*, and grains with $n_\ell^* < n < n_h^*$ should behave indifferent, since they can neither grow nor shrink. If the triple junction drag becomes very strong, it will eventually control the grain growth rate. In this case, the grain boundaries will become straight so that the grains assume the shape of polygons. This is because the curvature κ is directly proportional to Λ

$$n < 6:\ \kappa = \frac{\Lambda}{a}\sin\Theta(2\cos\Theta - 1)\exp\left[-\frac{2\Theta}{a}x\right]$$

$$n > 6:\ \kappa = \frac{\Lambda}{x_0}(1 - 2\cos\Theta)\exp\left(\frac{x}{x_0}\sin\Theta\right)$$

One can show that under these conditions a structure becomes stable if it is composed of only equilateral polygons [7] in contrast to the free-boundary-controlled kinetics where the stable structure consists only of hexagonal (six-sided) grains. Since the driving force does not depend on grain boundary curvature and, therefore, not on grain size, the grains will grow on average with constant velocity, i. e., the grain size increases in proportion to annealing time rather than to $t^{1/2}$ as in boundary-controlled motion.

This analytical mean field treatment is corroborated by computer simulations of grain growth via a vertex model, by which also the environment of a considered grain is taken into account [8]. The computer simulations confirm the theoretical prediction that for $\Lambda \to \infty$ the overall mean grain area S increases in proportion to the annealing time t, while for triple junction controlled motion $S \sim t^2$.

More importantly, however, it demonstrates that due to triple junction drag the growth rate dS/dt of a grain of topological class n is not uniquely defined anymore as predicted by the von Neumann–Mullins relation (Eq. (6)). Rather, dS/dt can assume – depending on the magnitude of Λ – any value between zero and a maximum growth rate which is given by the von Neumann–Mullins relation (Fig. 8). This behavior was recently confirmed by in-situ observations of triple junction motion in a SEM [3].

4. Conclusions

The motion of triple junctions was investigated in Al and Zn tricrystals. The experiments substantiate that triple junctions are crystal defects on their own with special properties. In particular, at lower temperatures, the triple junction mobility can be low which manifests itself in a deviation of the dihedral contact angles at triple junctions from the equilibrium angles. The effect of triple junctions can be expressed in terms of a criterion $\Lambda = (m_{tj} \cdot a/m_b)$, i. e., the ratio of junction and boundary mobility times the grain size. For

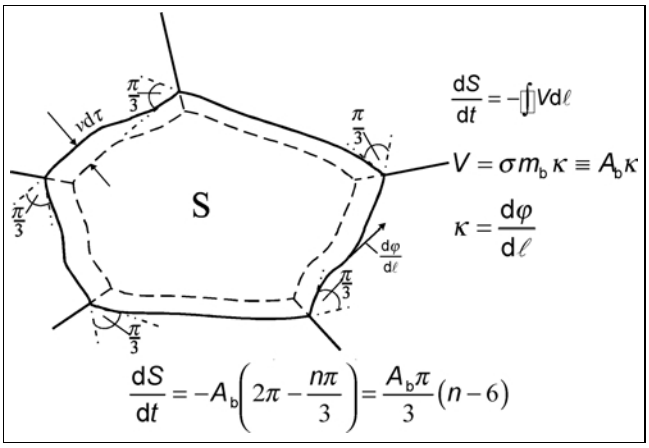

Fig. 6. Derivation of the von Neumann–Mullins relation.

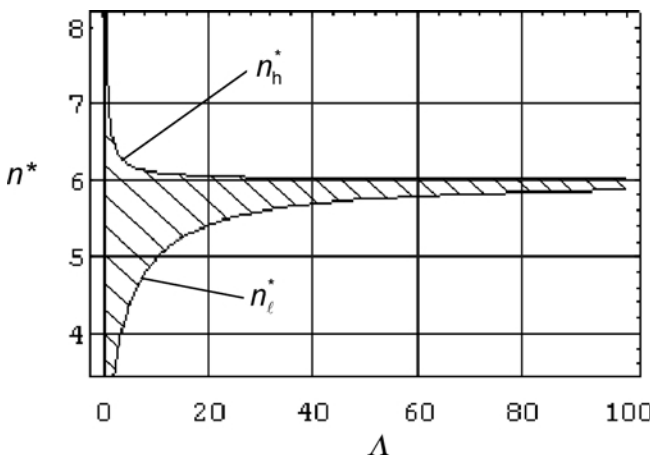

Fig. 7. The limiting topological classes for the triple junction configurations in Fig. 1a $\left(n_\ell^*\right)$ and Fig. 1b $\left(n_h^*\right)$.

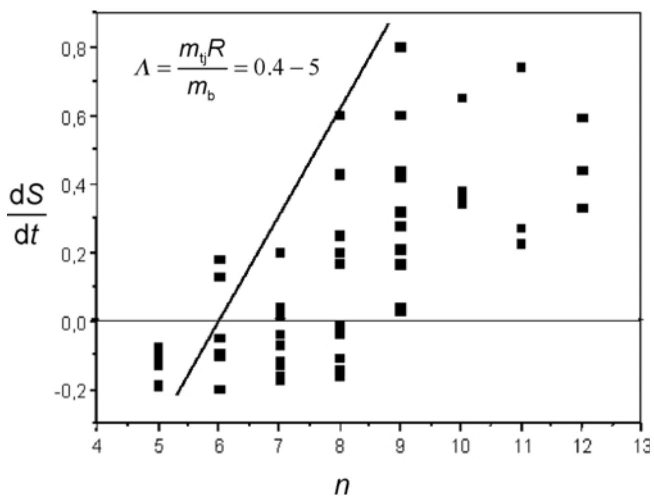

Fig. 8. The symbols denote the rate of area change of individual grains of a polycrystal computed by a vertex model assuming substantial triple junction drag. The straight line represents the von Neumann–Mullins approach.

small Λ, the triple junction strongly affects the kinetics of the connected boundaries. This effect is expected to be especially important for small grain sizes. We proved that low triple junction mobility also affects the von Neumann–Mullins relation of grain growth with the result that there is no unique relation between the growth rate and the number of sides of a grain anymore. For very low triple junction mobilities ($\Lambda \to 0$), the grains assume the shape of polygons, and the growth rate of grains does not change with annealing time. The theoretical concept is supported by computer simulations of grain growth and in-situ annealing experiments in the SEM.

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