PREFACE

Properties of materials are not mainly determined by their average chemical composition but substantially dependent on their microstructures. The large variety and complexity of microstructures typically encountered in engineering materials is strongly influenced by non-equilibrium mechanisms, e.g. recrystallization, which drastically changes the microstructure and its evolution. Microstructures that are highly non-equilibrium provide very often advantageous material properties, and not those microstructures that are close to equilibrium.

Only in the ideal case, a crystalline solid is a perfect, periodic structure and usually one can always find lattice defects in crystalline materials. Macroscopic properties of crystals are related to the entity of all lattice defects on a space scale that ranges from 10^{-10} m (impurity atoms) to 10^{0} m (sample surface), whereby its temporal evolution ranges from 10^{-12} seconds (dynamics of atoms) to 10^{8} seconds (creep, corrosion, or fatigue). It is one major aim of materials science to relate quantitatively macroscopic properties to microstructural features. This goal imposes the tasks to identify those lattice defects and to understand their collective static and dynamic behaviors that are responsible for specific macroscopic properties.

As a consequence, defects play an essential role in microstructure evolution, in particular two dimensional defects, i.e. internal surfaces between similar and dissimilar phases. Interfaces between equal phases but different crystallographic orientations are referred to as *grain boundaries*, which, apart from single crystals, exist in every crystalline material. They strongly influence the physical properties of a material, e.g. as a source or sink of point

defects. The comparison of the yield stresses of polycrystals and single crystals plainly makes their effect.

Grain boundaries have the unique property that they are able to react to exerted forces by a change of position. The motion of grain boundaries is the key phenomenon of recrystallization and grain growth; consequently, it seems reasonable to extract data on grain boundary motion from the temporal evolution of grain size during recrystallization or grain growth in polycrystals.

Although such data are useful to solve some specific problems, the relationship between grain boundary mobility and grain boundary structure, the effect of temperature, pressure, and impurity content on the motion of specific grain boundaries, the mechanisms of grain boundary migration, and other fundamental aspects of grain boundary motion cannot be studied by experiments on polycrystals, because in such experiments we only obtain information about the behavior of a large ensemble of different grain boundaries. Data, for instance the grain boundary mobility, are averaged over many different grain boundaries, like tilt and twist grain boundaries, low and high angle grain boundaries, or special and random grain boundaries as well as over many different rotation axes.
Moreover, polycrystals do not only contain grain boundaries, but also triple junctions, which have their own dynamic behavior and which can seriously affect the migration of grain boundaries.

In order to answer fundamental questions about grain boundary motion, and especially to clarify the mechanisms and the dependence of the mobility on different parameters, it is necessary to investigate the behavior of individual grain boundaries.

Mostly, grain boundary migration experiments have been conducted on curved grain boundaries, utilizing curvature as driving force. Since the structure of the grain boundary continuously changes along a curved grain boundary, the influence of the structure on mobility is not exactly known. In order to investigate the influence of grain boundary structure on the mobility, grain boundaries with a well known and constant structure are required. By using planar grain boundaries a constant structure can be obtained, but another driving force on the grain boundary is required to activate the grain boundary motion.

In 1952, Washburn and Parker have investigated planar low angle grain boundaries in Zn under the influence of an external shear stress and they were able to observe the continuous motion with polarized light in an optical microscope (Washburn and Parker 1952, Li et al. 1953, Bainbridge et al. 1954). Unfortunately, on the one hand, this observation technique does not work for cubic crystal structures, and on the other hand, any discontinuous

measurement of grain boundary motion is seriously affected by grooving. To overcome this problem one can use a method recently developed for continuous tracking of grain boundary motion in aluminum (Czubayko et al. 1995).

In the past, systematic investigations of grain boundary motion were confined to high angle tilt grain boundaries, while experimental data for the motion of low angle grain boundaries or twist grain boundaries are very scarce (Washburn und Parker 1952, Li et al. 1953, Sun and Bauer 1970, Viswanathan und Bauer 1973, Fukutomi und Horiuchi 1981). This is especially due to the fact that low angle grain boundaries as well as twist grain boundaries exhibit a small mobility, which complicates the already difficult measurement of grain boundary motion.

A driving force p on a grain boundary will force the grain boundary to move with the velocity v:

$$v = m \cdot p = m_0(c,\theta) \cdot \exp\left\{-\frac{\Delta H(c,\theta)}{kT}\right\} \cdot p \qquad (0.1)$$

with the mobility m and the driving force p. Grain boundary motion is usually a thermally activated process, therefore, the mobility m consists of a temperature-independent pre-exponential factor m_0 and the Boltzmann term, which contains the dependence on the temperature and the activation enthalpy for the grain boundary motion ΔH. The velocity depends on the chemical composition (concentration of impurities c), the grain boundary crystallography (angle of misorientation θ, rotation axis, grain boundary plane), the temperature T and the driving force p. For systematic investigations of grain boundary migration, it is necessary to know and to control all of these parameters.

As a consequence of the above mentioned problems, the current study will review a set of experiments to answer the following questions:

1. Is it possible to induce the motion of planar grain boundaries by application of a mechanical stress field?
2. What is the influence of the grain boundary structure on grain boundary motion?
3. What are the migration mechanisms of the stress-induced grain boundary motion?
4. Is there a difference between the behaviors of high and low angle grain boundaries and is it possible to determine the transition from low to high angle grain boundaries precisely?

In part I of this work, a short review about previous experimental results and models will be given to draw the attention to existing problems concerning grain boundary motion and to support the motivation for the following experimental investigations. In Chapter 2, results of experimental studies on *individual grain boundaries* are presented, which were performed to answer the questions given above.
In order to activate the motion of planar grain boundaries, a mechanical stress was applied to planar grain boundaries and the resulting stress-induced motion of different types of grain boundaries was investigated. The motion of symmetric tilt grain boundaries, asymmetric tilt grain boundaries, and twist grain boundaries was analyzed. In order to compare the results of the stress-driven motion with other types of grain boundary motion, experiments were carried out on the same grain boundaries but with curved grain boundary geometry without applying a mechanical stress field. In this case, the grain boundary moves due to a capillary force, which is, for instance, the driving force for grain growth. The comparison of the experiments is important for the discussion of the migration mechanisms.

In part II, theoretical concepts for the description of the interactions between mechanical stresses and grain boundaries will be presented. In Chapter 3, conditions and assumptions are introduced, which are the basis for the modeling and theoretical considerations of the interactions between mechanical stress fields and grain boundaries, and which give the mathematical description for the calculation of the driving force on grain boundaries in the experiments. Based on the experimental results of part I, in Chapter 4, the migration mechanisms of the different grain boundaries will be discussed and compared, especially the motions of curved and planar grain boundaries. Proceeding from the migration mechanisms, in Chapter 5, the term of the grain boundary mobility is introduced, in particular with regards to the questions, what the absolute grain boundary mobility exactly is and whether this absolute mobility exists. In Chapter 6, a theoretical consideration of the transition between low angle and high angle grain boundaries will be presented.

The conclusions based on the presented experimental results will clearly show that the dynamic behavior of individual grain boundaries can be influenced and changed by a mechanical stress field. The reaction of a grain boundary on the application of a mechanical stress defines the term *grain boundary mechanics*, which means all kinds of interactions between grain boundaries and mechanical stress fields. If it is possible to influence the behavior of individual grain boundaries by mechanical stresses, then we should also expect effects in case of grain boundary networks under the influence of mechanical stresses.

Therefore, in part III, the interactions between mechanical stress fields and grain boundaries in polycrystals will be discussed. In Chapter 8, the grain growth process is introduced and the common features of the grain growth are presented. The experiments on grain growth under an applied mechanical stress field are the subject of Chapter 9. Chapter 10 introduces a model, which is suitable to explain the experimental results of Chapter 9.

The microstructure in a polycrystal is strongly correlated to the properties of the material. Consequently, controlling the microstructure always means controlling the properties, which is the basic idea of grain boundary engineering, and which will be discussed in part IV. In Chapter 12, the main ideas of grain boundary engineering as well as an overview about the existing works in this field are presented. In Chapter 13, experiments will be introduced, which were performed to investigate the effect of grain boundary mechanics on the microstructure and the properties.

The current study is performed to illustrate how grain boundary mechanics can be used for microstructure control and for grain boundary engineering to improve materials properties and to evolve materials with optimized properties designed for particular applications.

PART I

GRAIN BOUNDARY PROPERTIES

INTRODUCTION TO PART I

In part I, results of experimental studies on individual grain boundaries are presented. In Chapter 1, a short review about previous experimental results and models will be given to draw the attention to existing problems concerning grain boundary motion and to supply the motivation for the following experimental investigations.

In all experiments the main question is, can a mechanical stress field influence the dynamic behavior of grain boundaries? To answer that question, different types of grain boundaries were investigated: symmetric tilt grain boundaries, asymmetric tilt grain boundaries, and twist grain boundaries.

The grain boundary motion, which could be measured in-situ by using X-ray diffraction on individually grown bicrystals, is described in Chapter 2. Generally, planar grain boundaries were used when a mechanical stress field was applied, so that the mechanical stress was the only driving force for the grain boundary motion. In order to compare the results of the stress driven motion with other types of grain boundary motion, experiments were carried out on the same bicrystals but with curved grain boundary geometry without applying a mechanical stress field. In this case, the grain boundary moves due to a capillary force, which is, for instance, the driving force for grain growth. The comparison of the experiments is important for the discussion of the migration mechanisms and the grain boundary mobilities in Chapter 4 and Chapter 5, respectively.

CHAPTER 1

REVIEW OF GRAIN BOUNDARY MOTION

1.1 Mathematical description of grain boundaries

In three-dimensional space, eight parameters are required to define unambiguously a grain boundary. These eight parameters can be divided into five macroscopic and three microscopic parameters. The five macroscopic parameters are three parameters for the orientation relationship, for instance the Euler angles φ_1, Φ, φ_2, and two parameters for the spatial orientation of the grain boundary by means of the normal unit vector of the grain boundary plane $\mathbf{n} = (n_1, n_2, n_3)$, with regard to one of the adjacent crystals. The five macroscopic parameters can be influenced externally. The three microscopic parameters are given by the three components of the translation vector $\mathbf{t} = (t_1, t_2, t_3)$ that characterizes the displacement of the two crystals with respect to each other. The translation vector will be forced by the crystals such that the total energy will be minimal; however, the vector **t** is not unambiguously defined as evident from computer simulations. The properties, in particular energy and mobility of a grain boundary, are, in principle, a function of these eight parameters.

Besides these eight parameters, which define the spatial position of the grain boundary, the grain boundary properties are influenced by other parameters. With regard to this, one can distinguish intrinsic parameters, such as impurities or other lattice defects, and extrinsic parameters, such as temperature or pressure.

Altogether, structure and properties of grain boundaries are dependent on many different parameters, whereby their investigation and description are complicated.

As already mentioned, the crystallographic orientation is different in two grains, which are separated by a grain boundary, but the crystal structure is the same in both grains. Therefore, it should be possible to make both grains coincide by a rotation. The general transformation consists of a rotation $\underline{\underline{R}}$ and a translation with the translation vector **t**:

$$\mathbf{r}^{(2)} = \underline{\underline{R}}\mathbf{r}^{(1)} + \mathbf{t}\,, \tag{1.1}$$

where $\mathbf{r}^{(2)}$ and $\mathbf{r}^{(1)}$ are the lattice vectors in grain 2 and grain 1, respectively. This rotation is an orthonormal transformation; consequently, the rotation matrix has to be independent of the type of description and of the choice of the coordinate system. This means, there is an infinite number of ways to prescribe a procedure of rotations that makes two coordinate systems coincide. In general, four ways are most convenient and, thus, most common to describe an orientation relationship:

- The three Euler angles: φ_1, Φ, φ_2. This description is widely used for an analytical treatment of rotations. It is standard for the calculation and representation of textures in terms of the three dimensional orientation distribution function (ODF).

- Miller indices: (hkl) [uvw]. The rotation is given by two crystallographic directions in coordinate system 1, which are parallel to the base vectors of coordinate system 2. It is commonly used when the crystal lattice is to be related to the specimen coordinate system, e.g. in a tensile sample with regard to the tensile axis or for rolling geometry with regard to rolling plane and rolling direction.

- Rodrigues vector: the vector points along the axis of the rotation, and its magnitude is the tangent of half the angle of the rotation.

- Axis and angle of rotation: θ [hkl]. It is easy to imagine and readily handled in stereographic projection. The notation is especially useful for the treatment of problems related to grain boundaries, as the type of grain boundary is commonly defined by the orientation of the grain boundary plane with regard to the axis of rotation.

In the following, we will usually utilize the last notation to describe the crystallography of grain boundaries.

1.2 Grain boundary motion

1.2.1 Energetic reasons for grain boundary motion

As already mentioned, grain boundaries have the unique property that they can react to exerted forces by a change of position. Grain boundaries move due to atomic processes in the vicinity of the grain boundary. Grain boundary motion can be divided into motion perpendicular to the grain boundary plane, in the following called *migration*, and motion parallel to the grain boundary plane, in the following called *sliding*. In general, grain boundary motion depends on a large number of parameters, e.g. the grain boundary structure (misorientation and inclination of the grain boundary plane), lattice defects (vacancies, dislocations, impurities), and extrinsic parameters such as temperature and, in particular, driving forces, which activate the process of motion.

In the following paragraphs, only the most important basics and features of grain boundary motion will be discussed, without any claim to completeness. For a deeper understanding of grain boundary motion, it is referred to some fundamental and extensive works in literature (e.g. Bollmann 1970, Gleiter and Chalmers 1972, Chadwick and Smith 1976, Balluffi 1980, Wolf and Yip 1992, Sutton and Balluffi 1995, Gottstein and Shvindlerman 1999).