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Meyer-Neldel Rule for the Kinetic Properties of Grain and Interphase Boundaries

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Abstract. The Meyer-Neldel rule (MNR) or compensation effect is observed in wide range of phenomena. MNR appears to be a fundamental property of many families of activated processes following an Arrhenius dependence on temperature. The kinetic properties of grain boundaries (GBs) and interphase boundaries (IBs) in solids depend strongly on their crystallographic parameters and obey the Arrhenius law. Therefore, the GBs form the families of objects where the Meyer-Neldel behaviour of thermally activated processes can be easily observed. The data on MNR for GBs in Al, Zn and Sn, GB triple joints in Al and Zn, and Sn–Ge IBs are presented. The data on "pressure NMR" for Sn tilt GBs and Ge–Sn twist IBs are presented as well. The thermodynamic model describing the activated state for GB diffusion and migration as a heterophase fluctuation is discussed. The values of compensation temperature for GB and IB diffusion and migration and GB triple joint migration are discussed using the idea of heterophase fluctuations.

Introduction

The compensation effect or Meyer-Neldel rule (MNR) [1] is observed in wide range of phenomena in physics, chemistry and biology [2-4]. Most frequently it was observed in case of thermally activated electrical conductivity [5-11]. MNR appears to be a fundamental property of many families of activated processes following an Arrhenius dependence on temperature,

$$\rho = \rho_0 \exp(H / kT) \qquad (1)$$

Here ρ is the absolute rate of a thermally activated process, ρ_0 is the preexponential factor, H is the activation enthalpy, and k is the Boltzmann constant. Commonly, by the evaluation of experimental data the activation enthalpy is determined from the slope H/k of an Arrhenius plot ln ρ vs. $1/T$.

It is frequently found that, when the activation enthalpy H is varied within a family of processes (for example, related chemical reactions), then the pre-exponential factor ρ_0 in (1) obeys the empirical relation

$$\rho_0 = \rho_{00} \exp(H / H_0). \qquad (2)$$

Here ρ_{00} is a constant, and H_0 is the Meyer-Neldel energy for the processes in question. Thus, the increase in the preexponential factor ρ_0 when H increases, Eq. (2), compensates for the decrease in the activation factor, Eq. (1), so that the processes actually take place at a rate larger than would be expected from the knowledge of H alone. The Eqs (2) and (3) imply the existence of a temperature T_C, called the compensation temperature, where all reaction rates ρ of the considered group of thermally activated processes are the same, i.e., the lines for the corresponding Arrhenius plots intersect at temperature T_C. The compensation effect or Meyer-Neldel rule is observed in wide range of thermally activated phenomena. It is relevant to the interpretation of experimental data for activated processes (e.g. diffusion, dark conductivity, thermoelectric power, field effect current, space-charge-limited current, thermal annealing of defects, and carrier emission from deep defects [12–14]). Therefore, it is important to understand its origin in detail.

The origin of the MNR is still debated. The more traditional theory [14] relates the MNR to the temperature induced shift of energy levels, and in particular that of the Fermi level, E_F. The shift of E_F with temperature ("the statistical shift ") is a consequence of the asymmetry in the density of states around it. Approximation of this shift with a linear temperature dependence leads to a discrepancy between the apparent activation energy and E_F, and a related discrepancy between the apparent and the microscopic [14] prefactors. Though this model is useful in explaining many of the MNR problems, it cannot explain the MNR behavior in annealing processes for atomic diffusion processes in crystalline materials [5].

It has been shown [12–16] that the MNR arises naturally for kinetic processes for which H is large in comparison with the energies of the excitations which contribute to the activation, as well as to kT. The exponential in Eq. (2) which is not at the origin of the effect, results, rather, from the entropy of combining multiple excitations (or fluctuations [17, 18]) in the thermal reservoir available for the kinetic processes. The Meyer-Neldel energy H_0, therefore, is expected to be of the order of the energy of the excitations in the reservoir, times a logarithmic correction term [12, 15, 16], which is frequently found to be of the order of unity. Among the phenomena which have been found to exhibit the MNR is diffusion in bulk solids [19–21]. In this case, the thermal reservoir contains the excitations of the lattice, i.e., phonons. In [5] the analytical model has been developed for the case of diffusion, where the circumstances appropriate for the NMR, i.e. the combination of multiple excitations in the realization of the kinetic process were treated. The molecular dynamic model [5] contains no exponential single-particle density of states, excluding such an explanation for the Meyer-Neldel behaviour observed [5]. Compensation, therefore, must arise from multiple excitations.

Important class of phenomena exhibiting the Meyer-Neldel behaviour is the kinetic properties of grain boundaries (GBs) and interphase boundaries (IBs). Already in the early experiments with the aid of individual GBs in bicrystals it has been shown that the spread of mobility m of moving GBs or their diffusion permeability $D_B\delta$ can reach several orders of magnitude [22–29]. The GB mobility and diffusion permeability depend hardly on the crystallographic parameters of GBs. Particularly, at certain misorientations, the lattices of both grains form the superlattice, namely the so-called coincidence site lattice (CSL). CSL in turn is characterized by the reverse density of coincidence sites Σ. Close to coincidence misorientations with low Σ the GB possess special structure and properties [30]. Usually, close to the coincidence misorientation with low Σ, GBs possess low mobility and diffusion permeability. Therefore, the GBs form the families of objects where the Meyer-Neldel behaviour of thermally activated processes can be easily observed. The experimental facts and thermodynamic model for GB Meyer-Neldel behaviour are presented here.

Thermodynamic model

In [31, 32] the thermodynamic model was proposed for the explanation of physical meaning of the Meyer-Neldel rule and compensation temperature. The preexponential factor in the Arrhenius relation, Eq. (1), consists of some geometrical constants ρ_1, a frequency factor ν and an entropy term exp (S/k) For a given material and kinetic process, $\rho_1\nu$ is constant and for convenience the dimensionless quantity $\rho/(\rho_1\nu) \equiv \tilde{\rho}_0$ can be considered instead, i.e.,

$$\tilde{\rho} = \exp\left(-\frac{G}{kT}\right) = \exp\left(-\frac{S}{k}\right)\exp\left(-\frac{H}{kT}\right) = \tilde{\rho}_0 \exp\left(-\frac{H}{kT}\right) \tag{3}$$

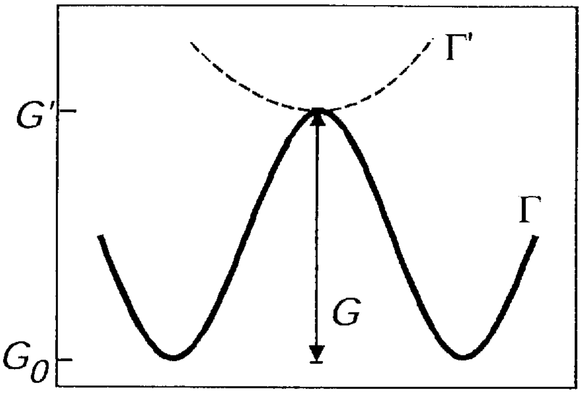


Fig. 1. Simplified correspondence of thermally activated state Γ and thermodynamically metastable state Γ'.

A linear relationship between H and $\log \tilde{p}_0$ is equivalent to the fact that the entropy of activation is linearly related to the activation enthalpy. This observed coupling of entropy and enthalpy of activation suggests that the activated state is not a random energy fluctuation, but a defined though metastable state which can be described by its respective thermodynamic functions. The transition from the stable ground state to the activated state corresponds to a first order phase transformation.In the GB or interphase boundary we can associate the activated state with a local change of the interface structure. In this sense we consider the saddle point configuration of the activated state Γ as a minimum free energy configuration of all potential metastable states for the given thermodynamic system (Fig. 1). The attainment of the activated state Γ' can be considered in this case as a first order displacive phase transformation. In this framework, the compensation temperature is the equilibrium temperature for such a virtual phase transformation.

The compensation relation, Eq. (2), can be easily derived under these conditions. As an example the GB mobility m can be considered, which is known to depend on GB structure and chemistry. Application of the Arrhenius relation (3) to the GB mobility m yields

$$\ln m = \ln m_0 - H_m / kT = \frac{S_m}{k} - \frac{H_m}{kT}, \tag{4}$$

where $S_m = k \ln m_0$ and H_m represent the activation entropy and enthalpy of GB mobility, respectively. Let the parameter λ denote some intensive structural or chemical specification, like angle of misorientation, composition, surface tension, etc. If λ changes slightly from the reference state λ_0, then S_m and H_m change accordingly

$$\ln m_0(\lambda) = S_m(\lambda)/k = \frac{1}{k}\left[S_m(\lambda_0) + dS_m/d\lambda\big|_{\lambda=\lambda_0}(\lambda-\lambda_0) + \ldots\right] \tag{5}$$

$$H_m(\lambda) = H_m(\lambda_0) + dH_m/d\lambda\big|_{\lambda=\lambda_0}(\lambda-\lambda_0) + \ldots \tag{6}$$

As S_m and H_m change only slightly, since free energy $G_m = H_m - T S_m$ is at minimum, a linear approximation is sufficient, and by solving Eqs. (5) and (6) for $\lambda - \lambda_0$ yields

$$\ln m_0(\lambda) = \frac{S_m(\lambda_0) - H_m(\lambda_0)/T_c}{k} + \frac{H_m(\lambda)}{kT_c}, \tag{7}$$

where

$$T_c = \frac{dH_m/d\lambda\big|_{\lambda=\lambda_0}}{dS_m/d\lambda\big|_{\lambda=\lambda_0}} = \frac{dH_m}{dS_m}\bigg|_{\lambda=\lambda_0} \tag{8}$$

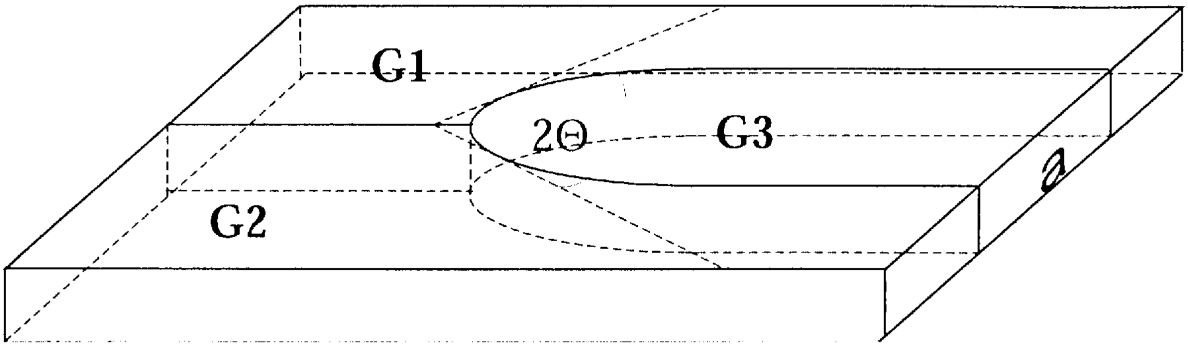


Fig. 2. Scheme of the tricrystal grown for the investigation of the triple junction motion in condition of a constant (capillary) driving force. *G1*, *G2* and *G3* – grains with diferent orientations; *a* – width of the grain *G3*; 2Θ – value of the vertex angle. Bicrystalline samples for the investigation of GB migration have the same geometry, but the GB between *G1* and *G2* is not present. The constant *a* ensures the constant (capillary) driving force.

is the compensation temperature, i.e., the temperature where ground state and activated state, or equilibrium phase and "barrier" phase are in equilibrium. This result implies that the barrier phase, i.e., the activated state, is a metastable phase closely related to the equilibrium state. It corresponds to a configuration of atoms with the smallest increase of potential energy with respect to the ground state. It seems obvious that equilibrium states occurring in the vicinity of the compensation temperature most easily satisfy this requirement. These conclusions are supported by the observation that the compensation temperature is often close to the equilibrium temperature of a near by phase transition [33]. Of course, when considering interface phenomena, potential metastable phases need not to be confined to bulk phases.

The simplified treatment given above delivers already the MNR, i.e., Eq. (2) and defines the compensation temperature. In [31, 32] a more sophysticated thermodynamic discussion of the activated state was given. This treatment is based on the assumption that the transition from the equilibrium state to the activated state is a first order phase transition. As a result, the generalized equation was obtained describing the linear relation between the enthalpy and entropy (or volume) of activation. In other words, there is a special, metastable, "barrier" phase, which is attained by a first order phase transformation at a critical temperature, pressure, etc. and which are referred to as compensation temperature, pressure, etc. Therefore, in analogy to the (temperature) compensation effect for constant pressure at variable temperature, there is also a (pressure) compensation effect for constant temperature at variable pressure. In other words, the activation entropy S^* is linearly related to the activation volume V^*. In fact, such relation was experimentally observed (Fig. 7) [35, 36]. Therefore, the MNR or compensation effect can be derived from equilibrium thermodynamics of the activated state which, in turn, can be associated with a potential metastable state. This metastable stete can be considered as a heterophase fluctuation [37].

Experimental

The bicrystals with individual GBs [22–29] and IBs [33, 34, 37–39] were grown using the modification of Bridgeman technique. For the investigation of GB diffusion the layer of duffusing agent is deposited on the surface of the bicrystal. The distribution of the diffusing agent is analyzed on the cross-section of the sample with the aid of radiotracer methods of electron probe microanalysis. For the investigation of GB mobility, the special geometry of bicrystal is used containing the elongated GB loop with parallel GBs (Fig. 2). This geometry provides the condition of constant (capillary) driving force for the GB migration. The velocity of moving GB can be

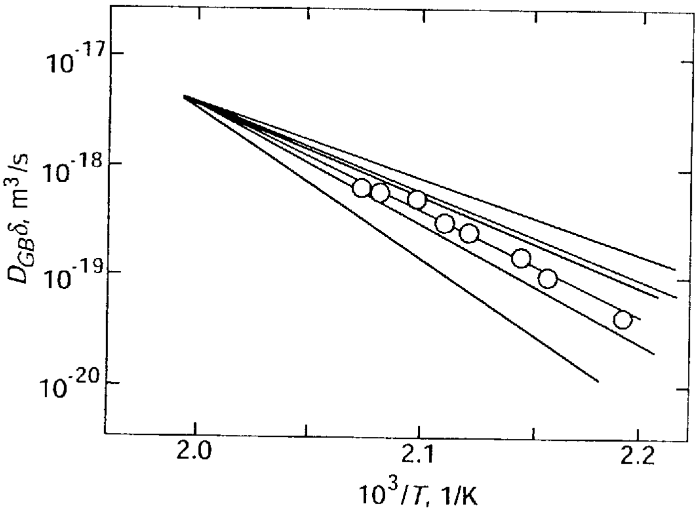


Fig. 3. Temperature dependence of the diffusional permeability $D_{GB}\delta$ of In in <001> twist Sn–Ge IBs in the temperature range 184–217°C [33, 39]. Misorientation angles (from top to the bottom): 2°, 3°, 14°, 39° and 40°. All experimental points (except IB with 14°) are omitted for simplicity. T_C = 220°C

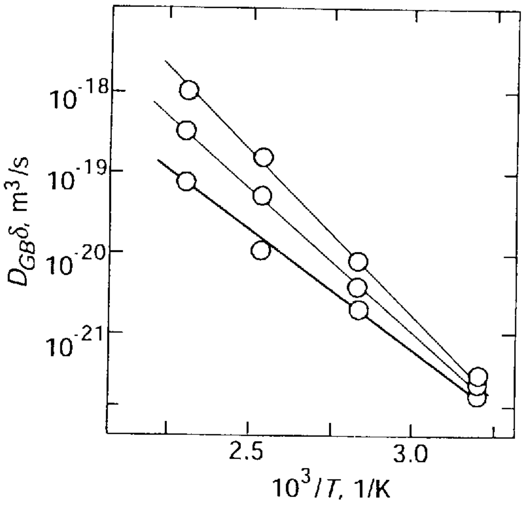


Fig. 4. Temperature dependence of the diffusional permeability $D_{GB}\delta$ of In in <001> twist Sn–Ge IBs in the temperature range 40–160°C [35, 39]. Misorientation angles (from top to the bottom): 2°, 6° and 0°. T_C = 27°C.

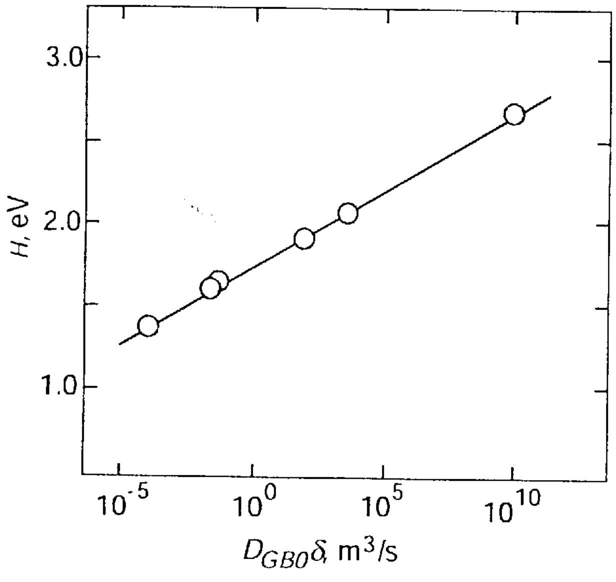


Fig. 5. Dependence of the activation enthalpy H for In diffusion along <001> twist Sn–Ge IBs on the preexponential factor $D_{GB0}\delta$ in the temperature range 184–217°C [33, 39]. T_C = 220°C.

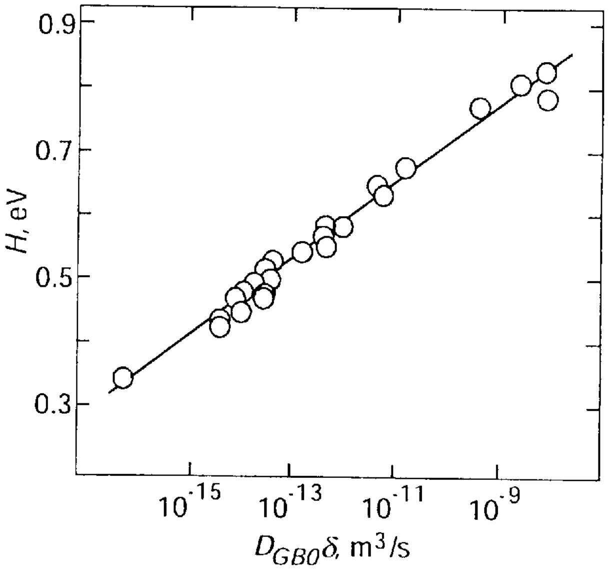


Fig. 6. Dependence of the activation enthalpy H for In diffusion along <001> twist Sn–Ge IBs on the preexponential factor $D_{GB0}\delta$ in the temperature range 40–160°C [35, 39]. T_C = 27°C.

measured *in situ* using the x-rays diffraction [45–47] or the polarized light if the material under investigation possesses the optical anisotropy (like, for example, zink) [25, 24]. The mobility of the GB triple junctions can be investigated using the tricrystals and modification of the method of constant (capillary) driving force (Fig. 2).

Results and discussion

In Figs. 3 and 4 the temperature dependences of the diffusional permeability $D_{GB}\delta$ of In in <001> twist Sn–Ge IBs are given for two different temperature ranges, namely 184–217°C (Fig. 3) and 40–160°C (Fig. 4) [33, 34, 39]. Diffusion permeability $D_{GB}\delta$ depends strongly on the IB misorientation angle. In the investigated Sn–Ge bicrystals the IB is perpendicular to the common four fold <001> axis of Sn and Ge grains. In case of the 0° misorientation angle the <010> axes of the Sn and Ge grains are also parallel. The comparison of Figs. 3 and 4 demonstrates that for In diffusion along Sn–Ge IBs two different compensation temperatures T_C exist, namely T_C = 220°C for the temperature range 184–217°C and T_C = 27°C for the temperature range 40–160°C. The respective MN-plots for activation enthalpy H and preexponential factor $D_{GB0}\delta$ are shown in Figs. 5 and 6.

In other words, the T_C in Fig. 3 lies above the temperature interval studied, and in Fig. 4 the T_C lies below the investigated temperature interval. In both cases the IBs having perfect crystallographic structure and low energy [37] possess the higher activation enthalpy of diffusion H in comparison with high-energy IBs [33, 34, 39]. However, in the temperature range of 184–217°C the IBs with high H have low diffusion permeability $D_{GB}\delta$ because the compensation temperature T_C lies above the temperature interval studied. Similar behaviour was observed by Zn diffusion along Al tilt and twist GBs in Al [26]. The compensation temperature in this system T_C = 500°C was well above the temperature interval studied [26]. *Visa versa*, in the lower temperature interval of 40–160°C the perfect IBs with high H possess high duffusion permeability $D_{GB}\delta$.

It is important to underline that the T_C = 220°C for the temperature range 184–217°C and T_C = 27°C for the temperature range 40–160°C are very close to the melting temperature of Sn and the temperature of phase transition αSn–βSn, respectively [48]. The MN-behaviour was observed also by the investigation of GB migration in Sn [35]. In that case the temperature interval studied was just under the melting point of Sn. The T_C obtained is very close to the Sn melting temperature as well [35]. These facts support the hypothesis on the nature of the activated state for GB and IB diffusion [31, 32, 36]. Obviously, the structure of the activated state in the Sn–Ge IBs is determined by the bulk phase transitions in Sn (melting and the αSn–βSn allothropic transformation). This hypothesis is additionally supported by the well known fact that at the meting temperature the diffusion coefficient has the same value in almost all solids which is close to the diffusion coefficicient in the respective melt [49].

Ge has much higher melting temperature than Sn [48]. Therefore, the kinetic properties of Sn–Ge IBs are determined in the temperature range 40–217°C mainly by the properties of Sn. Moreover, due to the very different physical properties, Sn and Ge form rather weak-bonded IBs. Therefore, the diffusion properties of the Sn–Ge IBs, including T_C, are governed mainly by the bulk properties of Sn and/or Sn–In solid solutions. Namely, the $D_{GB}\delta$ for In was measured in the concentration range between 0.5 and 6 at. % In. The crytical behaviour of $D_{GB}\delta$ was observed between 160 and 180°C which is due to the phase transition in the Sn–In solid solutions [50, 51].

Note, that the T_C = 500°C measured by Zn diffusion along Al tilt and twist GBs in Al [26] is rather far away from the melting point of Al [48]. The GB duffusion permeability $D_{GB}\delta$ was

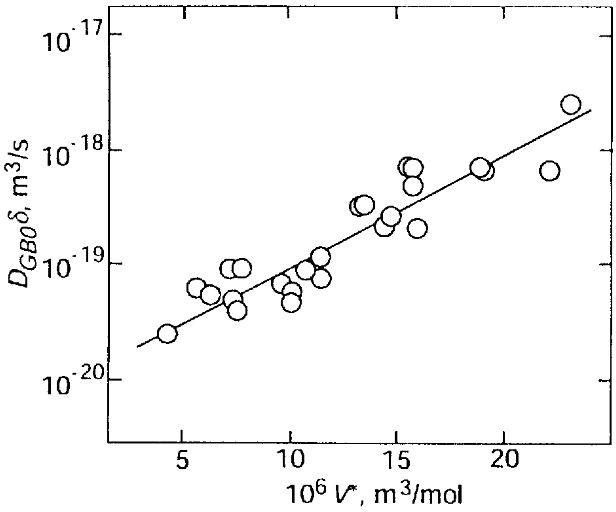


Fig. 7. Dependence of the product $D_{GB0}\delta$ at the atmospheric pressure on activation volume V^* at 160°C [34, 52]. P_C = 0.83 GPa.

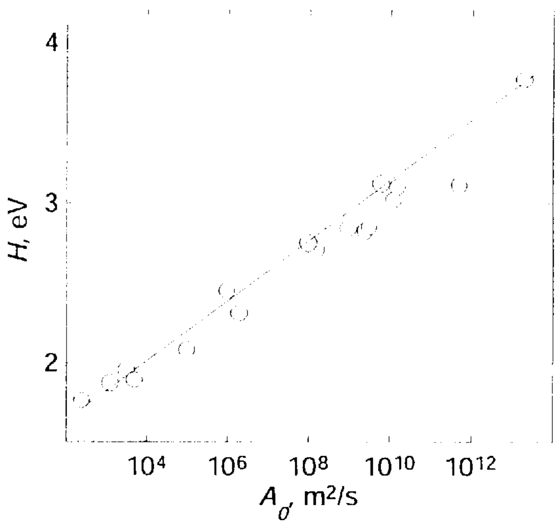


Fig. 8. The dependence of migration activation enthalpy H on the (reduced) preexponential mobility factor A_0 for <110> tilt GBs in Al [23, 40]. T_C = 590°C.

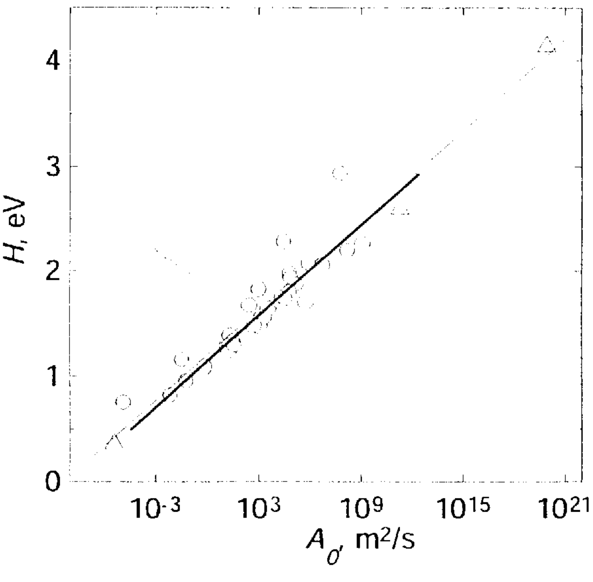


Fig. 9. The dependence of migration activation enthalpy H on the (reduced) preexponential mobility factor A_0 for <111> tilt GBs (circles, [23, 28, 40])and <111> GB triple joints (triangles, [41, 42]) in Al. For GBs T_C = 430°C. For GB triple joints T_C = 470°C.

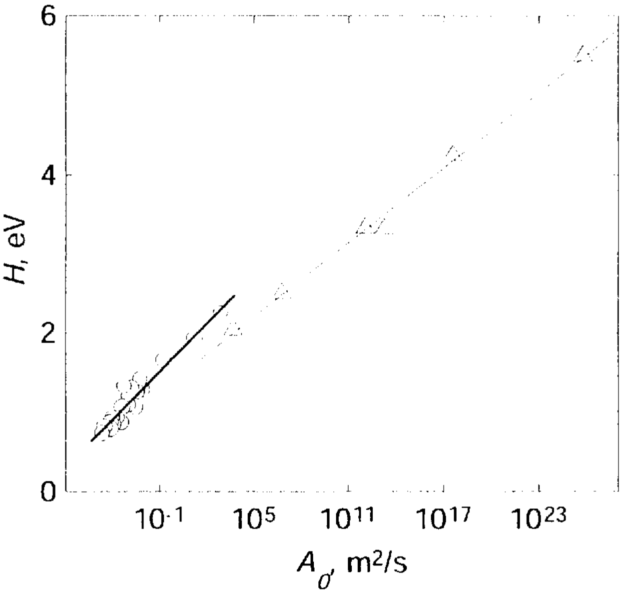
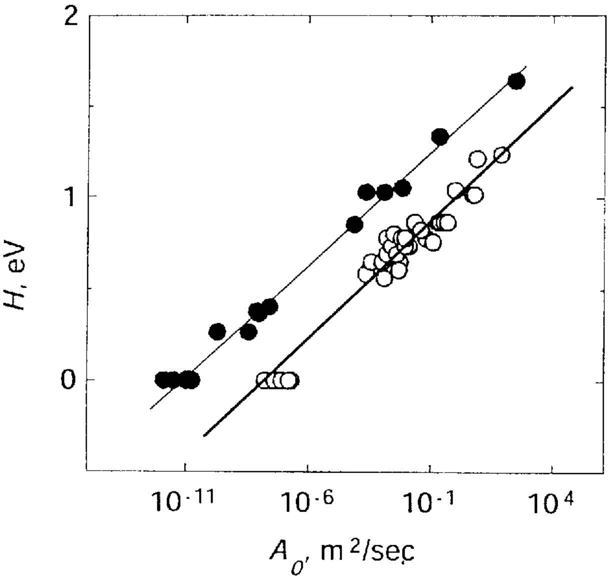


Fig. 10. The dependence of migration activation enthalpy H on the (reduced) preexponential mobility factor A_0 for <100> tilt GBs (circles, [24, 28])and triple joints (triangles, [41, 43]) in Al. For GBs T_C = 740°C. For GB triple joints T_C = 520°C.



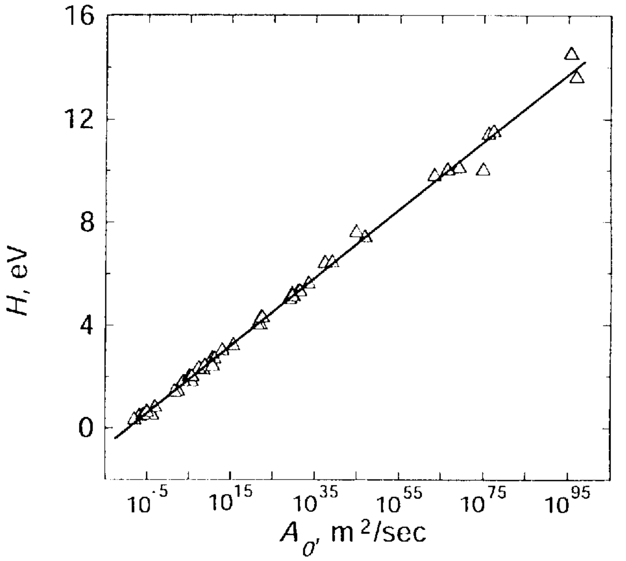


Fig. 11. The dependence of migration activation enthalpy H on the (reduced) preexponential mobility factor A_0 for $<11\bar{2}0>$ asymmetric twin GBs (full circles, [44]) and $<11\bar{2}0>$ tilt GBs (open circles, [25]) in Zn. For both cases T_C = 350°C.

Fig. 12. The dependence of migration activation enthalpy H on the (reduced) preexponential mobility factor A_0 for $<11\bar{2}0>$ and $<10\bar{1}0>$ tilt and general GB triple joints in Zn (this work). T_C = 420°C.

measured in the Zn concentration range between 0.5 and 15 at. % Zn [26]. Therefore, the T_C = 500°C can be attributed either to the solidus temperature or to the temperature of demixing phase transition in the concentrated Al–Zn solid solutions [48].

To the best of our knowledge, the so-called "pressure compensation effect" was for the first time observed in the In–(Sn–Ge) system [34, 52]. In Fig. 7 the dependence of the product $D_{GB0}\delta$ at the atmospheric pressure on activation volume V^* measured at the constant temperature of 160°C is shown. The slope of this log $D_{GB0}\delta$ (log V^*) plot yields the compensation pressure of P_C = 0.83 GPa. At this pressure all IBs studied have the same diffusion permeability. Similar MN-behaviour with increasing pressure was observed for Sn GB migration [35].

Generally, the GBs in metals are stronger bonded than Sn–Ge IBs. Therefore, it can be suggested that not only the bulk phase transitions but also the GB phase transformations can govern the structure of activated state and the resulting MN-behaviour. In Figs. 8 to 12 the data on the migration of GBs and GB triple joints are shown [23–25, 28, 40–44]. These experiments were performed with individual GBs and/or GB triple joints in conditions of the constant (capillary) driving force (cf. Fig. 2). The mobility A reduced to the same driving force (a in Fig. 2) was measured.

The pretty well MN-behaviour was observed in cases presented in Figs. 8 to 12. In case of <110> and <111> tilt GBs in Al the T_C = 590°C (Fig. 8) and T_C = 470°C (Fig. 10) lie well above the studied temperature range and can be rougly attributed to the melting point of Al [23, 24, 28, 40]. For the <110> and <111> tilt GBs in Al the same rule is fulfilled as for the IBs Sn–Ge, namely, the GBs with misorientation angles close to the low Σ coincidence misorientations possess the low GB energy, high activation enthalpy H and low mobility A. In case of <111> tilt GBs in Al the situation is more complicated [23, 28, 47]. In that case T_C = 430°C lies inside of the studied temperature range. This fact leads, particularly, to the complicated behaviour of the mobility of <111> tilt GBs close to the $\Sigma 7$ coincidence misorientation 38.2° <111> [47]. Namely, the maximum of mobility appears at 38.2° at low temperatures $T < T_C$. This maximum of A disappears at $T > T_C$. Such behaviour can be compared with the mobility of near-$\Sigma 17$ GBs in Sn below and above the GB phase transition "special GB – general GB" [53]. Therefore, the MN-behaviour of <111> tilt GBs in Al can be attributed to the GB phase transitions rather than to the bulk ones.

This hypothesis is supported also by the behaviour of $<11\bar{2}0>$ tilt GBs in Zn (Fig. 11) [25, 44]. Two groups of data are presented in Fig. 11. The open circles correspond to the bicrystalls with GB loops having constant width a (cf. Fig. 2). The full circles correspond to the "natural" individual twin plates obtained by the slight deformation of the $<11\bar{2}0>$ Zn single crystals. These twin plates were also perpendicular to the $<11\bar{2}0>$ plane of the sample. The values of activation enthalphy of Zn twins are definetely higher that those of of the $<11\bar{2}0>$ tilt GBs (Fig. 11). However, both groups of GBs possess the same T_C = 350°C. This temperature lies, like in case of <111> tilt GBs in Al, below the melting point and inside of the temeperature interval studied. Such behaviour can be explained by the possible GB phase transitions among the various constrained-CSL GB structures predicted in [54].

The experiments with individual GB triple joints have been started only recently [41–43]. However, already first data on GB triple joints reveal the MN-behaviour. In all cases studied the GB triple joints possess the much higher H values than those of comparable individual GBs (cf. Figs. 9 to 12). The intriguing high H values in Zn (see for example Fig. 12) force the investigator to think about possible mechanisms of GB triple joints migration. Both in case of <100> Al tilt GB triple joints (Fig. 10) and triple joints in Zn (Figs. 11 and 12) the compensation temperature differs from that of respective GBs. This fact and the high H values for triple joints support the suggestion that the triple joint migration is controlled by the processes in triple joint itself and not by the mobility of GBs forming the triple joint.

Conclusions

The MN-behaviour was observed by the investigation of migration of GBs and GB triple joints and by the studies of diffusion along GBs and IBs. The analysis of the compensation temperatures is based on the thermodynamic hypothesis that the activation statte can be treated as a heterophase fluctuation. This approach permits one to assign the different MN-behaviour of GBs, IBs and GB triple joints to the bulk phase transitions or to the GB phase tranasformations.

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