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M. Khandha, J. Cserti and V. Vitek

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

Univ of Pennsylvania
3231 Walnut Street, Suite 200
Philadelphia, PA 19104

8. PERFORMING ORGANIZATION
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We present a model for the anomalous increase of the yield stress exhibited by many L1₂ compounds. It is based on two thermally activated processes that describe respectively the pinning and unpinning of {101} screw dislocations in the {111} plane. The model explains all the important characteristic features observed in the anomalous regime. We discuss the applications of the model to Ni₃(Ga and Nb)(Al,Ta).

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THERMALLY ACTIVATED UNPINNING OF SCREW DISLOCATIONS IN THE ANOMALOUS REGIME IN $L1_2$ COMPOUNDS

M. Khantha, J. Cserti and V. Vittek
Department of Materials Science and Engineering
University of Pennsylvania
Philadelphia, PA 19104-6272.

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ABSTRACT

We present a model for the anomalous increase of the yield stress exhibited by many $L1_2$ compounds. It is based on two thermally activated processes that describe respectively the pinning and unpinning of $\{101\}$ screw dislocations in the (111) plane. The model explains all the important characteristic features observed in the anomalous regime. We discuss the applications of the model to Ni_3Ga and $Ni_3(Al,Ta)$.

INTRODUCTION

The anomalous increase of the yield stress with increase in temperature is well known in several $L1_2$ compounds and has been studied extensively. It is now recognized that the factors causing this behavior are also responsible for the many characteristic features in the anomalous regime such as (i) Strong orientation dependence of the yield stress and tension/compression (T/C) asymmetry [1-3]; (ii) A very low strain-rate sensitivity in the anomalous regime extending up to the peak temperature (T_p) [1, 4]; and (iii) A large discontinuity in the activation volume (v_a) at a temperature (T_c) well below T_p [4, 5]. While it has been known that a thermally activated mechanism obstructing the motion of screw dislocations in the (111) plane is the primary cause for the anomalous behavior, different approaches have been proposed to describe the unpinning or release of screw dislocations from the 'obstacles' that impede its motion. Many of the earlier models [1, 6] were based on a thermally activated process that 'pins' the screw dislocations but assumed an athermal process for the release. Such an approach could explain the feature (i) listed above as it is related to the details of the pinning mechanism but could not explain features (ii) and (iii) as they are related specifically to the thermally activated aspects of the release mechanism [7]. Both the pinning and unpinning mechanisms are assumed to be thermally activated in some recent models [8, 9] and agreement between theory and experiment has been demonstrated at certain selected temperatures and stresses. The starting point for all the models is common and is based on the thermally activated pinning mechanism proposed in the PPV model [6]. However, the models differ in the final dislocation configuration in the pinned state which then leads to differences in the unpinning mechanism as well. Recently, we have proposed a model [10, 11] which is also based on two thermally activated mechanisms and includes a simple unpinning process based on the concept of major breakaway that accounts for all the macroscopic characteristic features associated with the yield stress anomaly.

There are three main parts in our approach. The first describes a thermally activated process that results in the local pinning of screw dislocations in the (111) plane at random positions such that their movement in the glide plane is impeded. This part is qualitatively identical to the PPV model but includes some modifications in the saddle-point configuration in order to explain differences in the T/C asymmetry between binary and ternary compounds. Due to these changes, the activation enthalpy is somewhat different from that derived in the PPV model and has been described in [11]. It is based on the following description: Dislocation core transformations from a glissile to a sessile state are nucleated at random positions on mobile screw dislocations and result in the formation of a small cross-slipped segments on the (010) plane. The rest of the dislocation between the segments is bowed in the presence of stress.



Figure 1. The pinned state of the dislocation

The resulting configuration is shown schematically in Figure 1 in which the bow-out angle between two pinned segments is larger than the equilibrium bow-out angle determined by a single cross-slipped segment.

This however does not imply that the cross-slipped segment shrinks and disappears [12] since the path for unpinning a pinned segment is generally different and not necessarily related to the path which led to the nucleation of the segment.

In the second part, we consider the *major breakaway* (i.e., release from many pinning points and the subsequent return to the initial glissile state) of the dislocation via a thermally activated process by enumerating the various possible reaction paths for the release via different saddle-point configurations. The activation enthalpy for the release is obtained by choosing that path which gives the maximum rate of release while also enabling a major breakaway. We find that at low stresses/temperatures, when the mean separation between pinning points is large, the release from a single pin is rate controlling for major breakaway. However, as the stress (in effect, the temperature) increases, major breakaway becomes less likely after the release from a single pin due to re-pinning (or backward jumps) being more probable than further unpinning from the remaining pins. The release is then controlled by more difficult, but alternate, reaction paths that involve *multi-pin activation* such as the simultaneous activation at two or more pins. The change in the reaction path causes a discontinuity in the activation volume. As the temperature increases, eventually one obtains the condition that describes the transition from a dislocation that is pinned at discrete points to a dislocation that is pinned continuously all along its length. We identify the temperature at which such a transformation ensues to be the peak temperature.

The third part of the model prescribes the steady state of a system which is governed by two thermally activated mechanisms with very different waiting times. The thermally activated pinning and release mechanisms in steady state determine the resolved shear stress (RSS) on the (111) plane as a function of temperature for a given strain rate. We have tested the model in the entire anomalous regime by calculating the yield stress and the activation volume for different orientations in binary Ni_3Ga , Ni_3Al and ternary $\text{Ni}_3(\text{Al,Ta})$. The results compare well with observations including the prediction of the temperatures T_c and T_p . In addition, we predict at least one more discontinuity in the activation volume at a temperature, T_c' , between T_c and T_p . All the other important characteristics such as the orientation dependences, T/C asymmetry and the very low strain-rate sensitivity are also explained by this model [10].

THE UNPINNING MECHANISM

The concept of major breakaway and alternate reaction paths was first discussed in the context of internal friction phenomena [13-15] and we adopt a similar approach here. The basic features which distinguish major breakaway as opposed to the release from a single pin can be understood by considering a simple example of the thermally activated release of a dislocation with Burgers vector b and effective line tension τ pinned at two equidistant points separated by a distance L . In the presence of an applied stress σ , the dislocation bows out away from the pins, and let $y(x)$ be the displacement from the stress-free position in which the dislocation lies parallel to the x -axis. The equilibrium configurations of the pinned dislocation can be calculated by minimizing the energy within the line-tension model assuming a suitable dislocation-pin interaction energy, $U(y)$.

There are six non-equivalent equilibrium configurations [14] (see Figure 2) corresponding to the three displacement states possible at each pin, namely, pinned, saddle-point and unpinned states represented by 1, 2 and 3, respectively. A particular configuration is labelled as (ij) ($i, j = 1, 2, 3$) which denotes that the displacements at the two pins are in states i and j respectively. The configurations (11), (31) and (33) correspond to energy minima, (21) and (32) to saddle-points and (22) to a energy maximum.

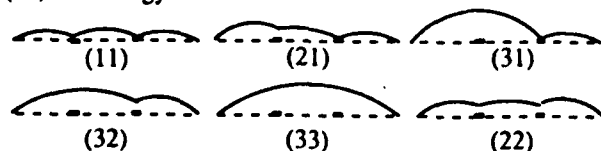


Fig. 2. The equilibrium configurations of a dislocation pinned at two points

The saddle-point state (21) involves activation at one pin while the saddle-point (32) involves simultaneous activation at both pins. Major breakaway corresponds to thermally activated transitions from state (11) to state (33) and this can occur via two distinct reaction paths: (i) (11)-(21)-(31)-(32)-(33) or, (ii) (11)-(32)-(33).

The (21) saddle-point is rate-controlling for transitions via the first path (since the subsequent passage through the state (32) is easier) while the (32) saddle-point controls the activation energy for the second path. We now examine which of these two alternatives provides the dominant contribution to the major breakaway by considering the activation enthalpy for transitions in the forward and backward directions (denoted by H_u and H_b respectively) for each path. The activation enthalpies are functions of the stress and the separation L . At low temperatures, when

pinning is infrequent and the value of L is large, major breakaway occurs only via the first path because the activation enthalpy for the second path is actually negative and represents an unphysical path because the simultaneous activation of two pins separated by a large distance is unlikely to be achieved. The path via (21) is, therefore, rate controlling provided the activation enthalpy for forward transitions leading to unpinning, $H_u(1)$, is smaller than the enthalpy $H_b(1)$, for the backward movement from (31) to (11) via (21) which leads to re-pinning.

As the value of L decreases (which is the case for the $L1_2$ alloys as the temperature increases), a situation when $H_u(1) \geq H_b(1)$ will ensue at a certain stress. When this happens, the release of the dislocation via the (21) path is no longer likely since the probability of re-pinning is larger than the probability of unpinning. However, it is precisely in this regime that major breakaway via the second path becomes feasible and in fact represents the dominant contribution for the following two reasons: (i) the enthalpy for forward jumps via the second path, $H_u(2)$, is equal to $H_u(1)$, which is the corresponding enthalpy for the first path and (ii) the enthalpy for backward jumps via the second path, $H_b(2)$, is much larger than $H_u(2)$, making re-pinning unfavorable for this path. Thus, while considering major breakaway, a transition from one dominant reaction path to another occurs when the release via the first path becomes unlikely due to re-pinning, i.e., when $H_u(1) \approx H_b(1)$.

This simple picture of release can now be generalized to the case of N pinning points [15]. Consider the configuration in which the dislocation has simultaneously broken away from $(m-1)$ adjacent pinning points and is in the saddle-point state at the m th pin ($m = 1, 2, \dots, N$) [15]. (The dislocation is assumed to be pinned with zero displacements beyond the m th pin). The displacements at the pins and the activation enthalpy can be expressed as functions of two dimensionless variables α and β given by $\alpha = (\sigma L^2 / 2\tau)$ and $\beta = (U_0 L / \tau b^2)$. Here U_0 represents the maximum value of $U(y)$ and α and β represent the combined effect of the stress σ and the separation L . Since the pinning points act as obstacles, the resisting force, $(-\partial U / \partial y)$, exerted by them on the dislocation during unpinning, must be positive everywhere. A simple form for $U(y)$ that exhibits such a behavior is the linear 'triangle-force' proposed in [13]. The interaction is of the type $U(Y)/U_0 = Y^2/pq$ for $0 \leq Y \leq p$, $U(Y)/U_0 = 1 - n(q - Y)^2$ for $p \leq Y \leq q$ and $U(Y)/U_0 = 1$ for $q \leq Y$; here p , q and n are constants and $Y (= y/b)$ represents the normalized displacement at the pin. We use this form of interaction in all our calculations with the same parameter values as in [13] ($n=1/8$, $p=1/3$ and $q=3$). The qualitative features of the unpinning process however, remain the same for all forms of $U(y)$ which rise monotonically from zero at $y=0$ to a certain maximum [15]. Let V_p , $V_{s.p.}$ and V_u represent the energies of the pinned (111...11), saddle-point (333...32) and the unpinned states (333...33) respectively. Using the triangle-force interaction, they can be expressed as

$$\frac{V_p}{U_0} = -\frac{\alpha^2}{\beta} \left[\frac{m+1}{6} + \frac{m^2}{m\beta+1} \right] ; \quad \frac{V_{s.p.}}{U_0} = (m-1) + (1-nq^2) - \frac{\alpha^2}{6\beta} (m^3+1) + \frac{[2\beta nq - (m+1)\alpha]^2}{2\beta \left(2\beta n - \left(\frac{m+1}{m} \right) \right)}$$

$$\frac{V_u}{U_0} = m - \frac{\alpha^2}{\beta} \left[m^2 + \left(\frac{m+1}{6} \right) \right] \quad (1)$$

The activation enthalpies for forward and backward jumps are given by $(V_{s.p.} - V_p)$ and $(V_{s.p.} - V_u)$ respectively. For large values of L , or more generally α , the controlling event for the unpinning of a dislocation is the release from a single pin ($m = 1$) while subsequent releases from adjacent pins require no further activation. When α decreases such that the backward jumps become as likely as the forward jumps, the major breakaway via this path becomes unlikely. The unpinning then proceeds by the next easiest path which is the simultaneous activation over two pins ($m = 2$) leading to major breakaway. However, when the backward jumps for $m = 2$ become easier than the forward jumps, the controlling path becomes simultaneous unpinning from three or more pins. A discontinuity in the activation volume $v_a = -\partial H_u / \partial \sigma$ occurs whenever there is a change in the dominant reaction path; v_a is usually much bigger for simultaneous activation at $(m+1)$ sites than for m sites.

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THE STEADY STATE

Let H_p represent the activation enthalpy for the pinning mechanism that results in the random nucleation of pinning points (or, segments of length ℓ_0) separated by a distance, ℓ , on average, along a segment of length L_s of a screw dislocation. We assume that after major breakaway, the dislocation glides freely on the (111) plane with a velocity V , travelling a distance d until enough pinning points have been nucleated on the unpinned segment that it again becomes sessile. The pinning process is more dominant than the unpinning process (although both are thermally activated) and is in fact the cause for the anomalous behavior. The rates of the two processes are therefore very different and hence a steady state cannot be achieved by simply equating their rates. If this is done, only the dominant process survives at long times while the weaker unpinning mechanism dies out completely in the steady state. In the present case, this would imply a complete transformation of screw dislocations into the sessile form. In order to attain a steady state in which both pinned and unpinned segments are present, a gain-loss rate equation must couple the dominant (pinning) and weak (unpinning) processes. The rate equations of predator-prey models, logistic equation and auto-catalytic reactions are examples in which strong and weak processes are coupled in a non-linear manner [16]. We adopt a similar but simpler approach here and use the characteristic time scale for the intermediate free-flight state between the pinned and unpinned configurations to write a linear coupled equation describing the rate of growth of pinned (or unpinned) segments at any time. In steady state, we approximate the average distance travelled in free flight d to the mean separation between pinning points, ℓ . We then obtain the steady-state condition (for details, see [10])

$$d = \ell = \sqrt[3]{(\ell_0 b V / v_0) \exp(H_p / 3kT)} \quad (2)$$

where v_0 is the Debye frequency, k is the Boltzmann constant and T is the temperature.

SCREW DISLOCATION MOTION AND THE STRAIN RATE IN THE ANOMALOUS REGIME

The rate of thermally assisted unpinning of screw dislocations moving on the (111) plane determines the strain rate which can be written as

$$\dot{\epsilon} = \dot{\epsilon}_0 \exp(-H_u / kT) \quad (3)$$

with $\dot{\epsilon}_0 = \rho b A (v_0 b / \ell) N$, ρ is the density of mobile dislocations, A the area swept by the unpinned dislocation and $N (\approx 1/\ell)$ the number of nucleation sites for unpinning per unit length of the dislocation. Since A is proportional to ℓ^2 (the proportionality constant may be large due to the major breakaway), $\dot{\epsilon}_0$ can be treated as a constant. The RSS on the (111) plane can be written as $\sigma_{pb} = \sigma_{pb}^0 + \sigma_{pb}^T$, where the superscripts 0 and T distinguish the athermal and thermal components of the stress, respectively. (In the following we identify σ_{pb}^0 with the low-temperature RSS.) The activation enthalpy H_u is calculated from equation (1) with L replaced by ℓ and σ replaced by σ_{pb}^T in α and β . For a given strain rate, the temperature dependence of σ_{pb}^T can be obtained from Equation 3 by finding the roots of the function $\Psi = [kT \ln(\dot{\epsilon}_0 / \dot{\epsilon}) - H_u]$ for the rate-controlling path chosen according to the criterion discussed above. For a given m , $H_u < H_b$ if $\alpha \geq \alpha_c = \sqrt{6\beta / (m+1)(m+2)}$ and thus this inequality determines when the simultaneous unpinning from m pins is the dominant process. The activation path for major breakaway changes from simultaneous activation at m pins to that at $m+1$ pins when $\alpha - \alpha_c$ changes sign from positive to negative. When the reaction path via (332) (i.e., $m = 3$) becomes unlikely, difficult paths via other saddle-point configurations such as (323), (213), etc., have been considered [14]. Since the major breakaway is unlikely to occur in this regime due to the high probability for pinning at high temperatures we approximate $d \approx \ell/3$ and calculate ℓ directly from the rate-equation [10].

According to equation (2) the separation of the pinning points decreases with increase in temperature and eventually their separation becomes comparable with their size, ℓ_0 , so that the dislocation is completely transformed into the sessile form all along its length. We identify the temperature at which this occurs as the peak temperature T_p . In the present context, this happens when all the configurations (saddle points and intermediate minima) become unstable except the states (11...1), (22...2) and (33...3). For true point-like pins, $\beta n < 1$ corresponds to a completely pinned dislocation. However, in the present case, the pinning points are actually segments of

length $2b - 3b$ and thus when $\beta n = 3-4$ the notion of discrete pins is no longer meaningful. Hence, we identify T_p by the condition that βn is in the range 3-4.

APPLICATION TO Ni_3Ga AND $\text{Ni}_3(\text{Al,Ta})$

The activation enthalpy H_p and the length ℓ_0 for the pinning process have been described in [11]. We choose the Burgers vector of the screw dislocation to be $2.52\text{\AA}$ in both the compounds. The separation between the Shockley partials which controls the constriction energy, W_c , has been determined using anisotropic elasticity. The values of APB energies on the (010) and (111) planes (in units of mJm^{-2}) have been taken as 200 and 237 in Ni_3Al containing 1 at.% Ta [17] and 20 and 100, respectively, in binary Ni_3Ga (an estimate); the CSF energy on the (111) planes has been estimated as 290, and 220, respectively. The energy gain, ΔE , in equation (2) of [11], has not been evaluated explicitly but we use an order of magnitude estimate, 0.01 (in units of $\text{Gb}^2/2\pi$) in $\text{Ni}_3(\text{Al,Ta})$ and 0.015 in Ni_3Ga . The factor, β_c , which takes into account the effect of impurities, introduced in [11], is zero in the binary compound and in $\text{Ni}_3(\text{Al,Ta})$ it is estimated to be -0.09 on the basis of the orientation dependence determined in [18]. The values of U_0 have been taken as 2.8eV in both Ni_3Ga and $\text{Ni}_3(\text{Al,Ta})$, and $\dot{\epsilon}_0 = 10^{12}\text{s}^{-1}$. For the free flight velocity, V , we use Leibfried's expression $V = 10\sigma_{pb}^{\frac{1}{2}}b^4 v_0/3kT$ [19]. The results are not sensitive to the actual values of U_0 , $\dot{\epsilon}_0$, ΔE and β_c but are more sensitive to the value of the CSF energy which is to be expected since small changes in the constriction energy and hence, H_p , produce large changes in ℓ .

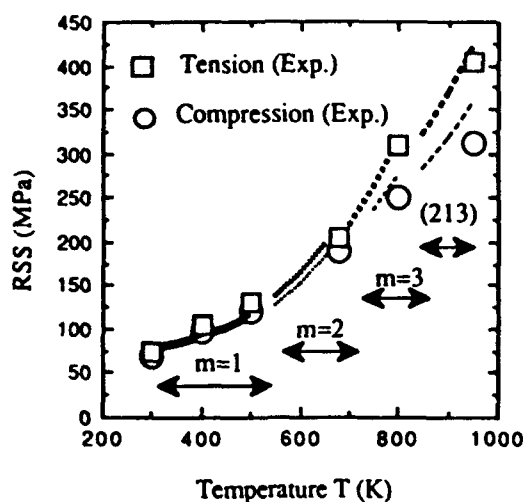


Figure 3. The calculated RSS vs T in Ni_3Ga .

Figure 3 shows the calculated temperature dependence of the RSS in the binary Ni_3Ga along with the experimental values [20] at $\dot{\epsilon} = 1.3 \times 10^{-3}\text{s}^{-1}$ for an orientation in which the ratio of the RSS on (111)[$\bar{1}21$] to the RSS on (111)[$\bar{1}01$] is 0.204. Following the experimental data σ_{pb}^0 has been chosen as 70 MPa. For this orientation the RSS in tension is higher than in compression which is borne out by the calculations. There are three distinct temperature regimes with different rate controlling paths. The $m = 1$ path is controlling from 300K to $\sim 500\text{K}$. Between 500K and 550K, when $\ell = 100b$, the $m = 2$ path starts to dominate and the activation volume exhibits a jump from $\approx 240b^3$ to $1000b^3$. This path gives the dominant contribution until 700 K. Between 700K and 750K, when $\ell = 45b$, there is another transition to the $m = 3$ path with a smaller jump in the activation volume.

Beyond 850K, paths such as (332) are no longer probable as H_p is less than H_u . Several alternate paths, (323), (213), etc., become possible [14] and it is difficult to decide unambiguously which one dominates. The value of the RSS is roughly the same for all the alternate paths but their activation volumes are slightly different. We show here the results obtained using the (213) saddle-point configuration. T_p determined from the condition that βn is in the range 3-4, is $\approx 950\text{K}$ agrees well with observations [20].

Figure 4 shows the calculated RSS in compression in ternary $\text{Ni}_3(\text{Al,Ta})$ for the [$\bar{1}23$] orientation at $\dot{\epsilon} = 1.3 \times 10^{-5}\text{s}^{-1}$ ($\sigma_{pb}^0 = 40\text{MPa}$) along with the experimental values [5]. The corresponding variation of the activation volume with temperature is shown in Figure 5 along with the apparent values measured in stress-relaxation experiments [5]. The first jump discontinuity at $T_c \sim 470\text{K}$ agrees well with observations. A second discontinuity is predicted to occur between 550 $\sim$ 650K resulting in a smaller jump magnitude. The orientation dependence, tension/compression asymmetry, the prediction of the peak temperature and the low strain-rate sensitivity are reproduced well by the model calculations [10].

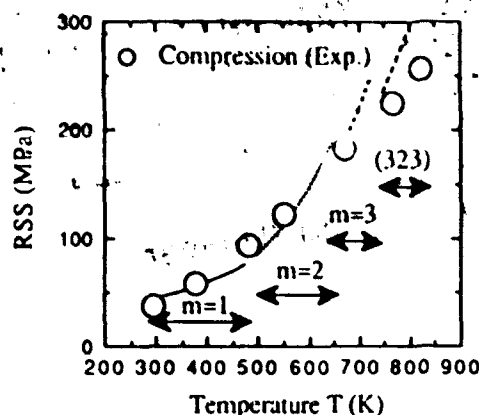


Fig. 4. The calculated RSS in compression vs T for the [1 2 3] orientation in $\text{Ni}_3(\text{Al,Ta})$.

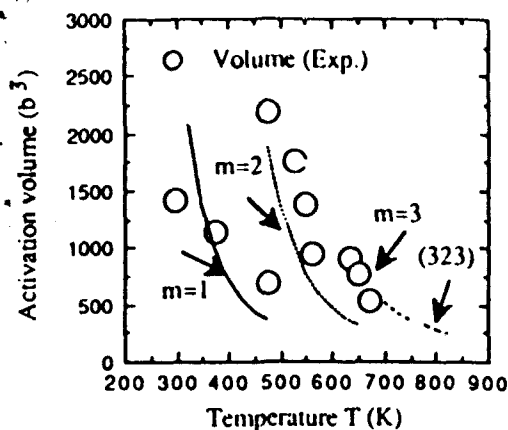


Fig. 5. The activation volume vs T in $\text{Ni}_3(\text{Al,Ta})$ for different dominant rate-controlling paths.

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