Constitutive modeling of $L_1_2$ intermetallic crystals

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Abstract

A dislocation model of the hardening of $L_1_2$ intermetallics is proposed. Hardening is presumed to be the net effect of point obstacles opposing the motion of dislocations. Two sources of point obstacles are considered: forest dislocations and cross-slip pinning. Forest hardening is assumed to be the only operative hardening mechanism for cubic systems. Octahedral systems are presumed to be subject to both forest hardening and hardening by cross-slip pinning. The rate of obstacle generation by the latter mechanism is taken to be proportional to the rate of cross-slip. The activation enthalpy for cross-slip is taken from the work of Paidar et al. (V. Paidar, D. P. Pope and V. Vitek, Acta Metall., 32 (3) (1984) 435).
Detailed comparisons between theory and experiment are given. The theory correctly predicts salient aspects of the behavior of $L_1_2$ intermetallics, including features which had previously been interpreted as indicative of “non-Schmid” yield behavior.

1. Introduction

Intermetallic compounds such as Ni$_3$Al are of interest in many applications owing to their low density, moderate ductility at room temperature, and their anomalous dependence of strength on temperature [1, 2]. For a sufficiently large offset strain, the conventional yield stress in uniaxial tension is observed to increase with temperature up to a peak and subsequently decrease at high temperatures. This anomalous behavior is nearly absent when very small offset strains are used to measure the yield stress, becomes accentuated as the offset strain is increased [3, 4], and then decreases again for very large offset strains [5]. The hardening rate of intermetallic single crystals also exhibits an anomalous dependence on temperature, namely, it increases at low temperatures while decreasing at high temperatures. Remarkably, Kuramoto and Pope [6] observe two peaks in the hardening slope of Cu$_3$Al at intermediate temperatures, an effect which has heretofore defied modeling.

An additional “anomaly” of $L_1_2$ intermetallic compounds is the tension-compression asymmetry in the uniaxial stress-strain curve [7, 8]. A common interpretation of this phenomenon [1] is to infer that the critical resolved shear stress for slip initiation (hereafter abbreviated as CRSS) is different in tension and compression, a so-called “non-Schmid” effect. Interestingly, the tension-compression asymmetry is absent at both low and high temperatures [7, 8]. Other forms of “non-Schmid” yield behavior, such as a dependence of the CRSS on the orientation of the loading axis, have been posited [9] in an effort to interpret the experimental record [10].

Many of the micromechanical processes underlying the behavior of $L_1_2$ intermetallics are presently well understood. Octahedral slip is known to be dominant at low temperatures, cubic slip at high temperatures, and both octahedral and cubic slip to coexist at intermediate temperatures [7, 8, 10]. Slip on octahedral systems is observed to be accompanied by cross-slip of screw segments into cubic planes, a process which is thermally activated and, consequently, is promoted by high temperatures [11]. Following cross-slip, dislocation segments become sessile and become “pinned”. Thornton et al. [3] suggested that the cross-slip pinning mechanism is responsible for the anomalous upturn in yield stress with temperature.

In an effort to explain the dependence of the CRSS on the orientation of the loading axis, Takuechi and Kuramoto [10] pointed out that the cross-slip rate (and hence the hardening rate) is likely to be promoted by a resolved shear stress acting on the cross-slip plane. In order to account for the tension-compression asymmetry, Lall et al. [7] noted that resolved shear stresses tending to contract the core of dissociated superdislocations have the effect of promoting cross-slip. Later,Paidar et al. [12] refined Lall et al.'s theory by accounting for the fact that the leading superpartial can alternatively dissociate within $\{111\}$ or $\{111\}$ planes.

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depending on the state of stress. Paidar et al.’s theory has been experimentally confirmed by Umakoshi et al. [8].

Despite this wealth of micromechanical evidence, a complete constitutive model of the hardening of L1₁ intermetallics which can be used in computation does not appear to be available at present. In this paper we endeavor to develop one such theory. We begin by questioning the notion that all the anomalies of L1₁ intermetallics are attributable to “non-Schmid” yield behavior. Indeed, as noted earlier, when very small offset strains are used to determine the yield point, the resulting yield stresses exhibit no discernible dependence on temperature and are free of anomalous effects [3, 4]. Only when sizable offset strains are employed and, therefore, hardening is allowed to take place, do anomalies arise in the purported yield behavior, such as the characteristic increase in strength with temperature. Furthermore, these anomalies become more pronounced with increasing offset strain. We take these observations as evidence that the true yield stress, i.e. the limit of the conventional yield stress as the offset strain tends to zero, is free of anomalous effects and that the anomalous “yield” behavior attributed to L₁₁ intermetallics is, in reality, a manifestation of their hardening behavior.

We shall therefore endeavor to develop a micromechanical theory of the hardening of L₁₁ intermetallic single crystals. The theory is constructed as follows. Hardening is presumed to be the net effect of point obstacles opposing the motion of dislocations. For monotonic loading, Cuitiño and Ortiz [13] have derived an explicit expression for the hardening rate of a slip system as a function of the density of point obstacles in that system. In the case of L₁₁ intermetallics, there are two main sources of point obstacles: forest dislocations and the cross-slip pinning mechanism described in the foregoing. In the absence of direct measurements, we resort to a simple geometrical derivation to relate the density of forest obstacles in a slip system to the dislocation densities in other systems. Then, we draw on the work of Gillis and Gilman [14] and Essmann and Rapp [15] to formulate the requisite equations of evolution for the dislocation densities as a function of slip activity.

Forest hardening is assumed to be the only operative hardening mechanism for cubic systems. By contrast, octahedral systems are presumed to be subject to both forest hardening and hardening by cross-slip pinning. The rate of obstacle generation by the latter mechanism is taken from the work of Paidar et al. [12], and is a function of the resolved shear stresses acting on the octahedral planes where the leading superpartial may dissociate, as well as of the resolved shear stress acting on the cross-slip cubic plane. Consequently, the rate of hardening of octahedral systems can depend markedly on the resolved shear stresses on other systems, which can be interpreted as a “non-Schmid” effect.

Detailed comparisons between theory and experiment are given in Section 4. The theory correctly predicts salient features of the behavior of L₁₁ intermetallics such as the anomalous dependence of strength on temperature, the equally anomalous temperature dependence of the rate of hardening, the tension-compression asymmetry of conventional yield (i.e. stress at a prespecified offset strain) at intermediate temperatures, the tension-compression symmetry of yield at both low and high temperatures, the dependence of the stress-strain curve on temperature and on the orientation of the axis of loading, and the patterns of slip activity as a function of temperature, deformation and the orientation of the loading axis. It bears emphasis that these features are not built directly into the theory, but are predicted by the theory.

2. General constitutive framework

The general structure of the constitutive equations describing the deformation of ductile single crystals is presently well established. For completeness, we summarize briefly the salient points of the theory. We restrict our attention to isothermal processes. More detailed accounts may be found elsewhere (for example ref. 13).

Under the conditions of interest here, the total deformation of a crystal is the result of two main mechanisms: dislocation motion within the active slip systems and lattice distortion. Following Lee [16], this suggests a multiplicative decomposition

\[ F = F^p F^c \] (1)

of the deformation gradient \( F \) into a plastic part \( F^p \), defined as the cumulative effect of dislocation motion, and an elastic part \( F^e \), which describes the distortion of the lattice. Following Teodosiu [17] and others [18–22] we shall assume that \( F^p \) leaves the crystal lattice not only essentially undistorted, but also unrotated. Thus, the rotation of the lattice is contained in \( F^e \). This choice of kinematics uniquely determines the decomposition (1).

Stresses in the crystal are induced by the lattice distortions described by \( F^e \). One might express this connection by, for instance, postulating a relation \( \tau(F^e) \) between the Kirchhoff stress tensor \( \tau \) and \( F^e \). A standard exercise shows that the most general form of this relation consistent with the principle of material frame indifference is

\[ \bar{\mathbf{S}} = \mathbf{S}(\bar{\mathbf{C}}^e) \] (2)
where \( \mathbf{S} = \mathbf{F}^{-1} \mathbf{t} \mathbf{F}^{e-T} \) is a symmetric second Piola-Kirchhoff stress tensor relative to the crystal lattice, and \( \mathbf{C} = \mathbf{F}^{e} \mathbf{T} \) is the elastic right Cauchy-Green deformation tensor. For most applications involving metals, a linear (but anisotropic) relation between \( \mathbf{S} \) and the elastic lagrangian strain \( \mathbf{E}^e = (\mathbf{C}^e - I)/2 \) can be assumed without much loss of generality. Higher-order moduli are given by Teodosiu [23].

From the kinematics of dislocation motion, Rice [22] derived the flow rule

\[
\mathbf{F}^p \mathbf{F}^{p-1} = \mathbf{L}^p = \sum_a \gamma^a \mathbf{s}^a \otimes \mathbf{m}^a
\]

where \( \gamma^a \) is the shear strain rate on system \( a \) and \( \mathbf{s}^a \) and \( \mathbf{m}^a \) are the corresponding slip direction and slip plane normal. In intermetallic compounds possessing the L1_2 structure, the potentially active slip systems are the twelve octahedral systems belonging to the family of [111] planes and [110] directions, and the six cubic systems of the type [001][110].

A simple calculation shows that the stress measure conjugate to \( \gamma^a \) is the resolved shear stress \( \tau^a \) acting on the plane of normal \( \mathbf{m}^a \) in the direction \( \mathbf{s}^a \). The resolved shear stress follows from, for example the Kirchhoff stress tensor \( \mathbf{t} \) through the relation

\[
\tau^a = \mathbf{s}^{aT} \mathbf{t} \mathbf{m}^a
\]

where one defines \( \mathbf{s}^a = \mathbf{F}^e \mathbf{s}^a \) and \( \mathbf{m}^a = \mathbf{F}^{e-T} \mathbf{m}^a \).

In view of the work conjugacy of the variables \( \tau^a \) and \( \gamma^a \), it is possible to interpret the resolved shear stress \( \tau^a \) as the driving force for the slip strain rate \( \dot{\gamma}^a \). For definiteness, we shall adopt the simple "viscosity law"

\[
\dot{\gamma}^a = \begin{cases} 
\dot{\gamma}_0^a \left( \frac{\tau^a}{g^a} \right)^{1/m} - 1 & \text{if } \tau^a > g^a \\
0 & \text{otherwise}
\end{cases}
\]

Here, \( m \) is the strain-rate sensitivity exponent, \( \dot{\gamma}_0^a \) is a reference shear strain rate, and \( g^a \) is the CRSS for slip initiation in system \( a \). Implicit in the form of eqn. (5) is the convention of differentiating between the positive and negative slip directions \( \pm \mathbf{m}^a \) for each slip system, whereupon the slip rates \( \dot{\gamma}^a \) can be constrained to be non-negative.

It should be carefully noted that we postulate the existence of a well defined yield point, or elastic limit, at \( \tau^a = g^a \) below which the system is inactive, \( i.e. \dot{\gamma}^a = 0 \). Thus, the CRSS \( g^a \) is identified with the zero "offset-strain" proportionality limit in the unloading-reloading curve, which must be carefully differentiated from the backextrapolation definition of the flow stress adopted in other theories. The zero offset-strain elastic limit of L1_2 intermetallics is free of anomalous effects [3, 4]. It is therefore appropriate to assume that the zero offset-strain elastic limit of a slip system is determined solely by the value of the resolved shear stress on that system, \( i.e. \) abides by Schmid's law. By way of contrast, micromechanical considerations (for example ref. 12) suggest that the rate of cross-slip from octahedral systems and hence their rate of hardening, depends on the resolved shear stresses acting on other systems as well. Therefore, in the present work, the non-Schmid effects characteristic of intermetallic compounds are presumed to emanate entirely from the micromechanisms of hardening of the crystal. A micromechanical model which supports this interpretation is developed in subsequent sections.

3. The hardening L1_2 intermetallic crystals

To render the preceding constitutive relations complete, an equation of evolution for the critical resolved shear stresses \( g^a \), or "hardening law", needs to be provided. The derivation of a hardening law for L1_2 intermetallic single crystals from dislocation mechanics is the focus of the remainder of the paper. We begin by considering the motion of dislocations within a slip system \( a \). This motion is the result of the intricate interplay between mobile dislocations, which are driven by the resolved shear stress \( \tau^a \), and obstacles. For single-phase crystals, secondary dislocations piercing the slip plane, or forest dislocations, constitute the principal source of obstacles. In the case of ordered intermetallic crystals with L1_2 structure, an additional source of obstacles is also operative in the octahedral systems, namely, the cross-slip pinning mechanism identified by Kear and Wilsdorf [11]. Both mechanisms result in an increase in the density of obstacles with deformation, thus contributing to the progressive hardening of the crystal. To estimate analytically the rate of hardening, we follow a two-step approach. Firstly, we characterize the "percolation"-like motion of dislocations through a random array of point obstacles, with the time variation of obstacle density presumed known. Secondly, we compute the rate of increase in obstacle density due to forest dislocation multiplication and the cross-slip mechanism.

3.1. Dislocation motion through random arrays of obstacles

Detailed numerical simulations of a dislocation line propagating through a random array of point obstacles have been carried out by Foreman and Makin [24, 25], and by Kocks [26]. An analytical treatment of the problem has been given by Cuitiño and Ortiz [13] within the statistical mechanical framework proposed by Ortiz and Popov [27]. In Kocks' model, all obstacles opposing the motion of the dislocation line are idealized as pinning points. Pairs of such points arrest dis-
locations, which require a certain threshold resolved shear stress $s$ to overcome the barrier, Fig. 1. For a given temperature, $s$ can be estimated from a line tension calculation (see, for example, the review of Kovács and Zsoldos [28]). The simplest such estimate gives

$$s = \frac{\alpha \mu b}{l}$$

(6)

where $\mu$ is the shear modulus, $b$ the length of the Burgers vector, $l$ the distance between pinning points, and $\alpha$ is a temperature-dependent coefficient. At room temperature, $\alpha = 0.3$ [29]. The variation of $\alpha$ with temperature $T$ is given by Seeger [30] and Saada [31] as

$$\alpha(T) = \begin{cases} \alpha_0 + (\alpha_c - \alpha_0)(1 - T/T_c) & \text{for } T \leq T_c \\ \alpha_0 & \text{for } T > T_c \end{cases}$$

(7)

Thus, $\alpha$ decreases linearly up to a critical temperature $T_c$ and remains constant at higher temperatures.

The motion of the dislocations through a random distribution of obstacles obeys a kinetic equation which was derived by Ortiz and Popov [27] using standard tools of non-equilibrium statistical mechanics. Cuitiño and Ortiz [13] were able to obtain analytical solutions of the governing kinetic equation for the case of monotonic loading and an arbitrary time variation of the density of point obstacles. These solutions determine the analytical form of the self-hardening curve. For randomly distributed obstacles in the slip plane, the result is

$$g^a = h^{aa} \gamma^a = h^a_c \left( \frac{g^a}{\tau_c^a} \right)^3 \left[ \cosh \left( \frac{\tau_c^a}{g^a} \right) - 1 \right]$$

$$h^a = \frac{\tau_c^a}{\gamma_c^a} \quad \tau_c^a = \alpha \mu b \sqrt{\pi n^a} \quad \gamma_c^a = \frac{b \rho^a}{2 n^a}$$

(8)

where $g^a$ is the critical resolved shear stress (in the sense of the elastic limit) of system $a$, $n^a$ is the density of point obstacles, $\rho^a$ is the dislocation density, $h^{aa}$ is the self-hardening modulus, $\tau_c^a$ is a characteristic of "flow" stress, $\gamma_c^a$ is a characteristic slip strain, and $h^a_c$ is a characteristic hardening modulus. The values of $\tau_c^a$ and $\gamma_c^a$ determine the location of the "bend" in the resolved shear stress-slip strain curve. In particular, $\tau_c^a$ correlates with the value of the flow stress determined by backextrapolation. As is evident from eqn. (8), $\tau_c^a$ and $\gamma_c^a$ are functions of the dislocation density $\rho^a$ and the density $n^a$ of point obstacles in the slip plane.

Remarkably, the hardening matrix predicted by the dislocation model is diagonal. Thus, the hardening law (8) conforms to the general structure suggested by Bassani and Wu [32] on the basis of their experimental data. By contrast, the conventional interpretation of latent hardening experiments [33] suggests that the hardening matrix is off-diagonally dominant. The conflict between these two views is only apparent, as it stems largely from the definition of flow stress adopted. If, as in the Pierce et al. [34] model, the emphasis is placed on $\tau_c^a$, which roughly corresponds to a backextrapolation definition of the flow stress, then the rate of hardening is computed from eqn. (8) as

$$\tau_c^a = \frac{\tau_c^a}{2 n^a} \rho^a$$

(9)

Because, in pure metals, the density of obstacles $n^a$ increases more rapidly with deformation in secondary systems than in the primary system, the resulting hardening matrix is off-diagonally dominant. If, in contrast, the rate of variation of the elastic limit with deformation is to be predicted, then a diagonal hardening matrix becomes appropriate.

The precise manner in which the dislocation model accounts for latent hardening effects in pure metals bears some emphasis. During the initial single slip regime, dislocation multiplication takes place predominantly on the primary slip system. Hence, the number of forest obstacles on the primary system remains relatively small, and the deformation proceeds by easy glide. At the same time, the number of obstacles on the secondary systems grows rapidly owing to dislocation multiplication on the primary system. This has the effect of raising the values of the characteristic flow stress $\tau_c^a$ on secondary systems. Consequently, while the CRSS $g^a$ remains small on the secondary...
systems, the reloading curve in a latent-hardening experiment simulation rises steeply above the primary loading curve, as observed experimentally [33]. A detailed comparison between the predictions of the dislocation theory and latent-hardening data has been given by Cuitiño and Ortiz [13].

3.2. Evolution of obstacle densities due to forest dislocation multiplication

In the foregoing discussion, the density of point obstacles \( n^\alpha \) in system \( \alpha \) was treated as a known function of time. Next we endeavor to derive equations of evolution predictive of the variation of \( n^\alpha \) with deformation. As noted earlier, in the case of L1\(_2\) intermetallic compounds of interest here, point obstacles arise from forest dislocations and, in octahedral systems, from the operation of the cross-slip pinning mechanism. In this section, we begin by addressing the first source of obstacles.

Following Franciosi and Zaoui [35], we postulate a linear dependence of the form

\[
n^\alpha = \sum_{\beta} \alpha^{\alpha \beta} \rho^\beta
\]  

between the density \( n^\alpha \) of point obstacles in slip system \( \alpha \) and the dislocation densities \( \rho^\beta \) in all slip systems. Experimentally determined values of the interaction matrix \( \alpha^{\alpha \beta} \) have been given by Franciosi and Zaoui [35] for f.c.c. crystals, and by Franciosi [36] for b.c.c. crystals. No similar experimental measurements for intermetallic crystals appear to be available at present. In view of this lack of data, we proceed to estimate the coefficients \( \alpha^{\alpha \beta} \) by recourse to a simple geometrical argument.

Let \( n^{\alpha \beta} \) denote the obstacle density introduced in system \( \alpha \) by dislocations in system \( \beta \). Evidently,

\[
n^\alpha = \sum_{\beta} n^{\alpha \beta}
\]  

Introduce an auxiliary reference frame with \( x_3 \) normal to the slip plane \( \alpha \) and \( x_2 \) coincident with the intersection between slip planes \( \alpha \) and \( \beta \), Fig. 2. Denote by \( \phi \) the angle subtended by dislocation segments in system \( \beta \) to the axis \( x_2 \). Assuming that the dislocation segments are randomly oriented, so that \( \phi \) is uniformly distributed from 0 to \( \pi \), the point-obstacle density contributed by all segments with orientations in the interval \( \phi, \phi + d\phi \) is

\[
dn^{\alpha \beta} = \rho^\beta \sin \phi \sin \theta^{\alpha \beta} d\phi
\]  

where \( \theta^{\alpha \beta} \) is the angle between planes \( \alpha \) and \( \beta \). The total density of point obstacles is, therefore,

\[
n^{\alpha \beta} = \int_0^\pi d\phi \rho^\beta \sin \theta^{\alpha \beta} = \frac{2}{\pi} \rho^\beta \sin \theta^{\alpha \beta}
\]  

Finally, we note that \( \theta^{\alpha \beta} = \arccos (m^\alpha \cdot m^\beta) \), so that

\[
\sin \theta^{\alpha \beta} = [1 - (m^\alpha \cdot m^\beta)^2]^{1/2}
\]  

Inserting eqns. (13) and (14) into eqn. (11) and comparing with eqn. (10), it follows that

\[
a^{\alpha \beta} = \frac{2}{\pi} - 1 - (m^\alpha \cdot m^\beta)^2)^{1/2}
\]  

which is the sought influence matrix. It bears emphasis that the influence matrix just derived is purely geometrical in nature and neglects the precise nature of the interactions between dislocations in the various slip systems. While for cases in which detailed measurements exist [35, 36] the influence matrix (15) does roughly predict the correct trends, it nevertheless oversimplifies matters by, for instance, neglecting the weak but non-vanishing interactions between coplanar systems [35]. Direct experimental measurements of the interaction coefficients \( a^{\alpha \beta} \) for L1\(_2\) intermetallic compounds remain highly desirable.

Equation (11) involves the dislocation densities \( \rho^\alpha \) on all slip systems. Processes resulting in changes in dislocation density include production by fixed sources, such as Frank–Read sources, breeding by cross-glide and pair annihilation (see ref. [28], for a recent review). The operation of fixed Frank–Read sources, however, usually stops after inducing a relatively small amount of plastic deformation. Consequently, production by fixed sources, while sometimes important during the early stages of plastic deformation, is quickly eclipsed by production due to cross-glide and can be safely neglected.

The importance of breeding by cross-glide as a dislocation generation mechanism was emphasized by
Johnston and Gilman [37, 38]. In this mechanism, screw segments migrate to a parallel plane by cross-slip, thus creating pairs of immobile jogs. The ends of the jogs act as the fixed points of single-ended plane sources. Theoretical [39] and experimental [37, 38] investigations suggest that the breeding rate due to cross-slip is proportional to the mean dislocation speed. This gives the relation

\[ b \dot{\rho}^a = \lambda \dot{\gamma}^a \]  \hspace{1cm} (16)

for the rate of dislocation generation due to cross-slip multiplication. The coefficient \( \lambda \) may be interpreted as the reciprocal mean free path between cross-slip events. The rate of pair annihilation is proportional to the probability of having two dislocations segments of opposite sign in a small neighborhood of each other, and thus may be expected to be roughly proportional to the dislocation density squared [40]. The average time between encounters is inversely proportional to the mean dislocation speed. Consequently, the rate of dislocation attrition due to pair annihilation can be expressed as [40]

\[ b \dot{\rho}^a = -R \rho^a \dot{\gamma}^a \]  \hspace{1cm} (17)

where \( R \) may be regarded as the mean radius of interaction for segment annihilation.

Combining eqns. (16) and (17), the total rate of change of dislocation density may be written in the suggestive form [14, 15]

\[ \dot{\rho}^a = \frac{\lambda}{b} \left( 1 - \frac{\rho^a}{\rho_{sat}} \right) \dot{\gamma}^a \]  \hspace{1cm} (18)

where \( \rho_{sat} = \lambda / R \) is a saturation density at which the rate of annihilation balances the rate of production. Equation (18) defines a linear ordinary differential equation for \( \rho^a \), the solution of which is

\[ \rho^a = \rho_{sat} \left( 1 - \left( 1 - \frac{\rho^a}{\rho_{sat}} \right) \exp \left( -\gamma^a / \gamma_{sat} \right) \right) \]  \hspace{1cm} (19)

where \( \gamma_{sat} = b \rho_{sat} / \lambda \) is a saturation shear strain, \( \rho^a_0 \) is the initial dislocation density in system \( \alpha \), and we have assumed that \( \gamma^a(0) = 0 \). It bears emphasis that relation (19) places the dislocation density \( \rho^a \) and the slip strain \( \gamma^a \) in one-to-one correspondence. Inserting eqn. (19) in eqn. (10) results in an explicit relation between the density of point obstacles due to forest dislocations and the slip strains in all slip systems.

3.3. Evolution of obstacle densities due to the cross-slip pinning mechanism

For cubic systems, forest dislocations are the sole source of point obstacles. Thus, inserting eqns. (10) and (19) into eqn. (8) determines the hardening behavior of these systems. By contrast, a second source of point obstacles on octahedral systems is the cross-slip pinning mechanism [11], which consists of the cross-slip of screw segments from [111] planes, where they are mobile, to [011] planes where they become immobile, Fig. 3. Each cross-slip event creates a pair of point obstacles on octahedral planes, thus contributing to the hardening of the crystal.

The rate of increase of obstacle density due to this mechanism is proportional to the number of cross-slip events per unit time. Therefore, introducing a mean free path \( L \) between cross-slip events yields the relation

\[ \dot{n}^a = \frac{\rho^a \dot{\gamma}^a}{L} \]  \hspace{1cm} (20)

where \( \dot{n}^a \) represents the rate of increase in obstacle density due to the cross-slip mechanism, and \( \dot{\gamma}^a \) is the mean speed of dislocations in system \( \alpha \). Using the well known expression \( \dot{\gamma}^a = b \rho^a \dot{\gamma}^a \) for the slip strain rate, eqn. (20) can be recast as

\[ \dot{n}^a = \frac{\dot{\gamma}^a}{bL} \]  \hspace{1cm} (21)

By virtue of the thermally activated character of the cross-slip mechanism, \( 1/L \) varies with absolute temperature \( T \) according to the Arrhenius law

\[ \frac{1}{L} = \frac{1}{L_0} \exp \left( -\frac{H}{kT} \right) \]  \hspace{1cm} (22)
where $L_0$ is the mean free path between cross-slip events at 0 K, $k$ is the Boltzmann constant and $H$ is the activation enthalpy.

Inserting eqn. (22) into eqn. (20) and integrating in time yields the expression

$$n^a = \frac{1}{bL_0} \exp \left( -\frac{H}{kT} \right) \gamma^a$$

(23)

for the contribution of the cross-slip pinning mechanism to the obstacle density in octahedral systems. The total obstacle density in these systems follows as the sum of the forest and cross-slip contributions, eqns. (10) and (23) respectively, with the result

$$n^a = \sum_\beta a^{\alpha\beta} \rho^{\beta} + \frac{1}{bL_0} \exp \left( -\frac{H}{kT} \right) \gamma^a$$

(24)

Equations (24) and (19), when inserted into eqn. (8), determine the hardening rate of octahedral systems. It is interesting to note that, while the contribution of forest dislocations to the obstacle density of system $a$ is proportional to the slip strain on $a$ itself, the rate of cross-slip, being thermally activated, increases sharply at high temperatures, thus increasing the rate of hardening of the crystal. At the same time, the strength of (111) point obstacles decreases with temperature according to eqn. (7). The competition between these two effects is responsible for the anomalous thermal effects characteristic of L1$_2$ intermetallics, see Section 4.1.

The activation enthalpy $H$ is required by a dislocation segment to cross slip from $\{11\}$ to $\{01\}$ planes is dependent on the configurations adopted by the segment before and after the cross glide event. Additionally, it’s value varies depending on whether the state of stress facilities or hinders the process. $\{11\}$ (101) superdislocations in L1$_2$ lattices dissociate into superpartials and adopt one of the following configurations. (a) Two 1/2 (101) superpartials separated by an antiphase boundary (APB). The cores of these superpartials are planar and further dissociated into 1/6 (211) and 1/6 (21) Shockley partials separated by a complex stacking fault (CSF). The resulting Peierls stress is low, which enhances the mobility of the dislocations. (b) 1/3 (211) and 1/3 (21) superpartials separated by an intrinsic stacking fault (SISF). In this case the cores are highly non-planar, which results in a high Peierls stress. Whether the first or the second configuration is preferred depends on the relative magnitudes of the APB and SISF energies. Materials such as Ni$_3$Al, Ni$_3$Ge and Ni$_3$Ga have a low ratio of APB to SISF energies, which renders the first dislocation structure energetically favorable. The situation is reversed for Pt$_3$Ga and Pt$_3$Al [12].

The cross-slip from $\{11\}$ to $\{01\}$ planes of a dislocation segment dissociated into superpartials bounding an APB has been studied by atomistic methods by Yamaguchi et al. [41] and Paidar et al. [42]. These studies suggest that the core of the leading superpartial can alternatively dissociate within $\{11\}$ or $\{11\}$ planes depending on the state stress. After cross-slip, the cores of the superpartials are no longer coplanar with the APB and, therefore, the dislocations become sessile. This accounts for the observed lack of mobility of the dislocation segments following cross-slip.

Since the superpartials are dissociated within $\{11\}$ planes before and after cross-slip, the activation energy for the process is diminished if the applied stress works to reduce the superpartial core width before cross-slip, and/or increases; the core width after metals, and was subsequently investigated by Paidar et al. [12] in the context of L1$_2$ intermetallics. Takeuchi and Kuramoto [10] additionally suggested that the activation energy is a decreasing function of the resolved shear stress on the cross-slip plane, which is sometimes known as the “cross-slip” effect.

Following Paidar et al. [12], a form of activation enthalpy for cross-slip which properly accounts for the core-width and cross-slip effects is

$$H = \frac{\mu b^3}{4\pi} (h + c + k(t_p - \kappa t_e)) - \frac{1}{(1/2)} - (\Gamma^{[001]}[\Gamma^{[111]}]) + |t_e[b/B]|^{1/2}$$

(25)

where $\Gamma^{[001]}$ and $\Gamma^{[111]}$ are the APB energies on [001] and [111] planes respectively, $h$, $c$, $k$ and $\kappa$ are dimensionless coefficients, and $B = \mu b^2/2\pi \Gamma^{[111]}$. In addition

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<th>TABLE 1. Material constants for Ni$_3$Al</th>
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<tr>
<td>Elastic constant $C_{11}$ (MPa)</td>
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<td>Elastic constant $C_{12}$ (MPa)</td>
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<tr>
<td>Elastic constant $C_{44}$ (MPa)</td>
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<tr>
<td>$\sigma^{[11]}$ (MPa)</td>
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<tr>
<td>$\sigma^{[00]}$ (MPa)</td>
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<tr>
<td>$\gamma^{{00}}$ (s$^{-1}$)</td>
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<tr>
<td>$\alpha$</td>
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<tr>
<td>$b$ (m)</td>
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<tr>
<td>$\rho^{{00}}$ (m$^{-2}$)</td>
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<tr>
<td>$\rho_{av}$ (m$^{-2}$)</td>
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<tr>
<td>$\alpha_0$</td>
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<tr>
<td>$\Gamma^{[111]}$ (J m$^{-2}$)</td>
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<tr>
<td>$\Gamma^{[001]}$ (J m$^{-2}$)</td>
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<tr>
<td>$h$</td>
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<tr>
<td>$c$</td>
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<tr>
<td>$k$</td>
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<td>$\kappa$</td>
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<td>$L_0$ (m)</td>
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$\tau_{pe}$ and $\tau_{se}$ denote the resolved shear stresses on the primary and secondary octahedral planes in the direction of the edge component of the Burgers vector, $\langle 111 \rangle \langle 121 \rangle$ and $\langle 111 \rangle \langle 121 \rangle$ respectively, $\tau_{so}$ denotes the resolved shear stress on the cross-slip cubic system $\{010\} \langle 101 \rangle$, and one writes $t = \tau / \Gamma^{|111|} / b$. Numerical values of the constants for Ni$_3$Al are given in Table 1. Details of the derivation of eqn. (25) and of the physical significance of the constants may be found in the original references of Padian et al. [12] and Escaig [43].

It bears emphasis that, owing to the form, eqn (25), of $H$, the rate of cross-slip and the rate of hardening in an octahedral system depend on the resolved shear stresses acting on other systems, which can be loosely interpreted as a "non-Schmid" effect. It should be carefully noted, however, that in the present theory the onset and rate of slip of a system are, for a given state of hardening of the crystal as described by the values of $g^a$, fully determined by the resolved shear stress on that system through eqn. (5).

**Summary of constitutive relations**

\[
\gamma^a = \begin{cases} 
\gamma_0 (|t^a / g^a|^{1/m} - 1) & \text{if } t^a > g^a \\
0 & \text{otherwise}
\end{cases}
\]

\[g^a = h^a \gamma^a\]

\[h^a = h_0 \left( \frac{a^a}{\tau^a} \right)^3 \cosh \left[ \left( \frac{\tau^a}{g^a} - 1 \right) \right]\]

\[h^a = h_0 \left( \frac{a^a}{\tau^a} \right)^3 \cosh \left[ \left( \frac{\tau^a}{g^a} - 1 \right) \right] \]

\[\gamma^a = \frac{b \rho^a}{2 n^a}\]

\[n^a = \begin{cases} 
\sum \alpha^a \rho^a & \text{for cubic systems} \\
\sum \alpha^a \rho^a + \frac{1}{bl_0} \exp \left( - \frac{H}{kT} \right) \gamma^a & \text{for octahedral systems}
\end{cases}\]

\[\rho^a = \rho_{sat} \left[ 1 - \left( 1 - \frac{\theta_0}{\rho_{sat}} \right) \exp(-\gamma^a / \gamma_{sat}) \right]\]

**4. Comparisons between theory and experiment**

In this section we compare the predictions of the dislocation model with selected experimental data. We restrict our attention to Ni$_3$Al single crystals, and to the uniaxial tension-compression test. The theory correctly accounts for key aspects of the experimental record, such as the anomalous increase and subsequent drop in the yield stress with increasing temperature, the widely differing patterns of slip activity observed at low and high temperatures, the initial increase and subsequent decrease of the hardening rate with temperature, and others. It bears emphasis that all of these features are predicted by, and not built into, the theory. The good overall agreement with the available observational evidence demonstrates the predictive nature of the theory.

The numerical procedure adopted for the integration of the constitutive equations has been described in detail elsewhere [13]. The constitutive update is fully implicit, with the active systems (i.e., those systems for which $\tau^a > g^a$ at the end of the time step) determined iteratively. Loading and boundary conditions are accounted for by recourse to a simple 8-noded cubic finite element. The element is aligned with the loading axis, and the two loaded faces of the cube are constrained to remain parallel to each other and perpendicular to the loading axis. These boundary conditions simulate a stiff loading device in which the clamps are prevented from rotating. All stress-strain curves are reported in terms of nominal stress and engineering strain. For ease of reference, the constitutive relations are collected at the end of Section 3. The values of the material constants employed in the calculations are listed in Table 1.

**4.1. Yield stress vs. temperature**

Several definitions of "yield stress" are commonly adopted in constitutive theories and in experimental studies, most notably the true yield stress or elastic limit, the conventional yield stress defined as the stress at a prespecified offset plastic strain, and the flow stress defined by backextrapolation. These definitions are not equivalent, and they quantify different physical phenomena. Thus, the true yield stress signals the point of inception of plastic flow. The conventional yield stress depends on the offset plastic strain adopted, and reflects not only the strength but also the hardening characteristics of the crystal. The conventional yield stress reduces to the true yield stress in the limit of a zero offset strain. Evidently, the influence of hardening on the value of the conventional yield stress increases with increasing offset strain.

The uniaxial tension data reported by Thornton et al. [3] and Mulford and Pope [4] on Ni$_3$Al, reveal that the conventional yield stress is nearly independent of temperature for very small values of the offset strain, e.g., $10^{-6}$. For sufficiently high offset strains, the conventional yield stress attains a maximum at a critical temperature $T_{max}$, an effect which is considered "anomalous" and becomes more pronounced with increasing offset strain. We take these observations as...
strong evidence that the true yield stress is independent of temperature and that the anomalous "yield stress" behavior characteristic of Ni$_3$Al is, in actual reality, a manifestation of its hardening behavior. Note, however, that the anomalous yield behavior becomes ameliorated at very large offset strains, an effect which is clearly evident in Fig. 6. This trend is also consistent with experimental observations [5].

In keeping with these observations, we have taken the initial value $g_0^0$ of the CRSS in all slip systems to be a constant independent of temperature, with the initial CRSS $g_0^{[111]}$ of cubic systems exceeding that $g_0^{[001]}$ of octahedral systems by a factor of 2. Despite the constancy of $g_0^0$, the dislocation model predicts the anomalous behavior observed experimentally, Fig. 4. The good qualitative agreement between the theoretical predictions shown in Fig. 4 and the experimental data of Thornton et al. [3] and Mulford and Pope [4] is noteworthy.

The precise manner in which the dislocation model accounts for the anomalous "yield" behavior is consistent with the available observational evidence [3, 4, 6–8, 10] and may be summarized as follows. Owing to the higher value of the initial yield stress in cubic systems, octahedral systems are the first to yield at all temperatures. For small offset strains, the constancy of the initial CRSS in all systems results in an equally constant value of the computed conventional yield stress independent of temperature, Fig. 4.

At low temperatures, $T \ll T_{\text{max}}$, the rate of cross-slip is relatively small and hardening is governed by the forest mechanism. In crystals oriented for single slip, however, no appreciable dislocation multiplication takes place on secondary systems during the early stages of deformation. Consequently, the rate of forest hardening is small and deformation proceeds by easy glide. Under these conditions, the stress–strain curve is ostensibly flat, and the recorded conventional yield stresses are fairly insensitive to the choice of offset strain, Fig. 4. In the range spanned by the usual offset strains, the stress levels attained do not suffice to cause yielding in the cubic systems, which remain inactive.

As the temperature is raised, cross-slip is promoted by thermal activation and the crystal hardens at an increased rate by the cross-slip pinning mechanism. This results in higher levels of stress and the cubic systems become active at increasingly smaller strains. Indeed, near the critical temperature octahedral and cubic systems are predicted to exhibit comparable levels of activity for deformations in the usual range of offset strains, in keeping with experimental observations [7]. Under conditions of multiple slip, the active systems harden each other rapidly by the forest mechanism. This effect, in conjunction with the increased hardening by cross-slip pinning, accounts for the upturn in the conventional yield stress with increasing temperature.

At high temperatures, $T \gg T_{\text{max}}$, cubic systems yield at small strains and, because they do not harden by cross-slip, they quickly become the dominant active systems. While forest hardening still remains operative, the effectiveness of this mechanism diminishes with temperature as a result of the progressive loss of strength of point obstacles predicted by the Seeger–Saada relation [30, 31]. Beyond the critical temperature, this effect becomes dominant and the conventional yield stress drops with increasing temperature.

### 4.2. Tension–compression asymmetry

An additional "anomaly" of intermetallic compounds such as Ni$_3$Al is the asymmetry exhibited by their uniaxial stress–strain curve depending on whether the loading is tensile or compressive [7, 8, 10]. A common interpretation of this phenomenon [1] is to infer that the initial critical resolved shear stresses $g_0^0$ are different in tension and compression, a so-called "non-Schmid" effect. But, as mentioned previously, initial critical resolved shear stresses appear to be free of anomalous effects [3, 4]. In view of this fact, we shall favor the view that the tension–compression asymmetry is a consequence of the hardening characteristics of the crystal. Because hardening begins to operate immediately after yield, it may mask the elastic limit and affect conventional measurements of the yield stress, even when small offset strains are adopted. In this manner, a feature of the hardening behavior may be mistaken for a property of the initial CRSS.

Figure 5 shows the predicted conventional 0.2% yield stress as a function of absolute temperature for
behavior is evident from the figure. It should be noted, however, that this anomalous behavior is not introduced directly into the formulation as a temperature dependent yield condition but is predicted mechanistically by the theory, as discussed in Section 4.1.

Also evident in Fig. 5 is the tension–compression asymmetry, which is a direct consequence of the asymmetry of Paidar et al.'s expression accounts for the fact that cross-slip is strongly influenced by the resolved shear stress acting on the cross-slip plane, as well as by the value and sign of the resolved shear stresses acting on the planes where the superpartials are dissociated. When the loading axis is close to the [001] direction, Fig. 5(a), the tensile yield stress is predicted to be higher than the compressive yield stress. The reverse situation is obtained when the loading axis is centered on the standard triangle, Fig. 5(b), or is near the [111] direction, Fig. 5(c). These predictions are in agreement with the experimentally observed trends [8].

Also in agreement with experiment [8] is the computed convergence of the tensile and compressive yield stresses at low and high temperatures, Fig. 5. Behind this convergence lies the fact that, both at low and high temperatures, forest hardening is dominant. Indeed, at low temperatures cross-slip, being thermally activated, occurs relatively infrequently. At high temperatures, the crystal deforms predominantly by slip on the cubic systems, which are subject to forest hardening only. However, forest hardening is insensitive to the sign of the load and, consequently, the tension–compression asymmetry is eliminated in the absence of hardening by cross-slip pinning.

4.3 Stress–strain curves

Copley and Kear [5] and Umakoshi et al. [8] reported experimentally measured uniaxial tension stress–strain curves for Ni₃Al single crystals. For low symmetry orientations of the loading axis, at low and high temperatures the curves exhibit an initial stage of low and nearly linear hardening, followed by a second stage of rapid hardening. By contrast, near the peak temperature “parabolic” hardening is observed. Parabolic hardening is also observed at low and high temperatures when the loading axis is close to a high symmetry direction [44].

This behavior is readily explained within the present theory. For low symmetry loading axes, forest hardening results in the stage I–stage II stress–strain response characteristic of f.c.c. ductile single crystals (see Cuitiño and Ortiz [13], for detailed simulations). In Ni₃Al, at temperatures much lower (higher) than the peak temperature deformation proceeds primarily by slip on octahedral (cubic) systems without significant hardening by cross-slip pinning, and thus stress–strain curves with a stage I–stage II structure are obtained, Fig. 6.
The initial stage I of low hardening corresponds to deformation by single slip. When stresses rise sufficiently for multiple slip to begin, the rate of forest hardening increases sharply and stage II sets in. At high temperatures, however, the stage II of hardening is less pronounced owing to the progressive reduction in the strength of point obstacles. When the loading axis coincides with a direction of high symmetry, multiple slip takes place from the outset and the stage I of hardening is absent, in agreement with Aoki and Izumi's [44] observations. Near the peak temperature, cross-slip pinning gains in importance and parabolic hardening is obtained, Fig. 6.

4.4. Hardening rate

The hardening slope in a uniaxial tension test exhibits a variation with temperature similar to that exhibited by the conventional yield stress, i.e., it increases with temperature in the range $T < T_{\text{max}}$ and decreases with temperature in the range $T > T_{\text{max}}$. Remarkably, Kuramoto and Pope [6] observe two peaks in the hardening slope of Cu$_3$Al at intermediate temperatures, an effect which had heretofore defied modeling.

The variation of hardening slope with temperature predicted by the dislocation model is in keeping with these observations, Fig. 7. Shown in the figure are the average slopes over a prespecified range of deformation computed from the uniaxial stress–strain curves collected in Fig. 6. While the values of the hardening rate so computed are somewhat sensitive to the choice of sampling strain interval, the overall trend is not. At low temperatures, the stress–strain curve exhibits an initial stage I of low hardening, which results in low values of the hardening slope. As the peak temperature is approached, parabolic hardening due to cross-slip pinning sets in, and an upturn in the average hardening slope results. Near the peak temperature, several competing effects shape the stress–strain curve. Cross-slip pinning on octahedral systems becomes increasingly dominant with temperature, which promotes parabolic hardening. At the same time, cubic systems gradually increase their activity and, because they are only subject to forest hardening, give the stress–strain curve an increasingly linear shape. Finally, the strength of point defects decreases with temperature, which diminishes the effectiveness of all hardening mechanisms. These competing pressures result in the vacillating double-peak variation of the average hardening slope near the transition temperature, Fig. 7. In the range $T > T_{\text{max}}$, the progressive loss of strength of point obstacles becomes the dominant effect and the hardening slope decreases monotonically with temperature.

4.5. Slip activity

Experimentally, octahedral slip is known to be dominant at low temperatures, cubic slip at high temperatures, and both octahedral and cubic slip to coexist at intermediate temperatures [7, 8, 10]. Plots of slip activity of the most active cubic and octahedral systems as a function of axial strain at low, intermediate and high temperatures are shown in Fig. 8. The loading axis is centered within the standard triangle, which initially favors single slip. At $T = 300$ K, the deformation proceeds by single slip on the octahedral (001) [110] system up to considerable axial strains. By contrast, at the intermediate temperature $T = 600$ K, the initial stage of single slip on (001) [110] is much shorter, and is followed by double slip involving the cubic system (001) [110]. Following the onset of double slip, the activity in the octahedral system is sharply diminished and the cubic system becomes dominant.

![Ni$_3$Al](image1)

![Ni$_3$Al](image2)

Fig. 6. Theoretical uniaxial stress–strain curves at different temperatures.

Fig. 7. Hardening rate in uniaxial tension vs. temperature for two offset strains.
Fig. 8. Slip activity vs. uniaxial deformation in the most active octahedral and cubic systems, at (a) 300 K, (b) 600 K, and (c) 900 K.

These trends are accentuated at $T = 900$ K, at which temperature the cubic system (001) [110] quickly becomes overwhelmingly dominant following a short stage of single slip on (001) [110].

Acknowledgments

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References