Influence of temperature on twinning dominated pop-ins during nanoindentation of a magnesium single crystal

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Abstract
The present paper examines the temperature sensitivity of tensile twinning in a magnesium single crystal during nanoindentation of the prismatic plane. High temperature indentations from 25 °C to 250 °C were employed on a well polished magnesium single crystal \([10-10]\) plane \([10-10]\) plane. For a indentation curve displaying a pop-in, a single twin was seen on the sample surface using Atomic Force Microscopy (AFM) imaging. For indentations that produced no pop-in, no twinning was observed. We thus conclude the pop-in arises from a twinning event in the present case. With increasing temperature, the mean pop-in load (measured from 200 repeat indentations of each testing temperature) drops markedly. This is interpreted by the thermal activation of nucleation of lattice dislocations, which immediately trigger a twinning event. Thermal activation analysis yields activation energies that are consistent with this idea. With increasing temperature the pop-ins became deeper and the twins, after further indentation, showed more growth. It is likely that non-basal slip is activated in the stress concentrations that arise during twinning and the thermal activation of this slip accounts for the observed temperature effects. It is concluded that in interpreting the temperature sensitivity of twinning stresses, any associated lattice dislocation activity must be considered.

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1. Introduction
Mechanical twinning is frequently observed in magnesium and its alloys. The common \(\text{hcp} \{1012\}\) ‘tensile’ twinning mode (the only twin mode that will be discussed in the present work) is readily activated due to its relatively low critical resolved shear stress (CRSS) \([1,2]\). With increasing temperatures, the activity of twinning tends to drop \([1,3-11]\). This is generally understood to be due in magnesium to the increased ease of non-basal slip modes which operate preferentially at higher temperatures (e.g. \([3,4]\)). The transition point is obviously affected by the temperature sensitivity of twinning, which is typically considered to be weak or even negligible. However, the literature on this contains conflicting results. In one case higher \([1012]\) twin fractions have been seen with increased temperature \([12]\) and, in another two cases, an increase in \([1012]\) twin CRSS with temperature has been reported \([4,10]\). It has also been observed that \([1012]\) twins formed at higher temperatures can be thicker than those formed at lower temperatures \([13]\). There is a need for further insight into how temperature impacts on the nucleation and growth of twins in magnesium.

Our experimental understanding of how twin nucleation and growth contribute to this stress has been mediated by statistical analysis of \textit{post-mortem} microstructures \([14-17]\), studies of bulk material behaviour \([18-21]\), \textit{in-situ} transmission electron microscopy (TEM) \([22,23]\), micro-pillar testing \([4,24-27]\) and nanoindentation \([28-31]\). Nanoindentation is unique in that it provides the potential to probe twin forma-

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tion in-situ in partly constrained single crystal material volumes characterized by very low initial dislocation densities. This provides opportunity to examine the role of lattice dislocations on twin formation and the relaxation of the twin back-stress. These are phenomena likely to impact on the temperature dependency of twinning.

Nanoindentation is marked by sudden displacement bursts (pop-ins) when performed on metal surfaces prepared to ensure the local dislocation density is low [32–37]. This is often ascribed to the rapid multiplication and migration of lattice dislocations, particularly following a dislocation nucleation event. Morris et al. [37,38] and Phani et al. [39] developed a statistical model based on residual dislocations underneath the indenter to explain the size-dependency of the pop-in stresses. However, when the indented area is defect-free, the shear stress at pop-in approaches the theoretical stress and homogeneous nucleation of dislocations can occur [37,40–42]. Schuh et al. [43,44] also have been able to rationalize the temperature sensitivity of pop-in events in terms of dislocation nucleation. In line with this, Catoor et al. [45] have interpreted pop-in events seen in magnesium in terms of lattice dislocation activity. They employed the Indentation Schmid Factor (ISF) defined by Li et al. [33] to infer likely active slip modes. In complementary work, Somekawa et al. [46] found that lattice dislocation motion could explain the pop-in phenomena they observed in their magnesium simulations and experiments.

However, during indentation of Mg and its alloys it is difficult to avoid activating twinning. Nanoindentation tests on Mg alloys with different grain sizes [47,48] revealed [1012] twins that increased in activity with grain size. Twins have been observed [49,50] at the sides of indents when indentaion was performed normal to basal and prismatic planes. Profuse twinning during nanoindentation has also been observed using various indenter geometries [51,52]. Multiple [1012] twinning events were observed around impressions made using larger indenters (1 mm in diameter) [53] or preforming higher load indentations [52]. Nayyeri et al. [54,55] carried out indentations nearly parallel to the c-axis of pure Mg and revealed depth-dependant [1012] twinning events during both loading and unloading regimes. In the work of Somekawa et al. [46] quoted above, they note that their study did not reveal twinning possibly due to the low indentation depths employed.

Twinning typically occurs in a rapid burst of strain and thus can be expected to give rise to pop-ins during nanoindentation. Pop-ins seen during nanoindentation of the {0001} plane of pure Mg have been interpreted in terms of individual twinning events [55]. Indented prismatic [1010] surfaces have been seen to consistently display individual twins following a single pop-in event [29–31]. This has been verified using three-dimensional Electron Backscatter Diffraction (EBSD) technique [30]. Twin thickness measurements on [1010] surfaces have been found to be in reasonable agreement with what one would expect if the pop-in displacement was accommodated by the twinning shear [29]. Thus, during indentation of [1010] planes, pop-ins appear to be highly sensitive to the nature of single twinning events [28–31]. The present study will validate this proposition and examine the temperature sensitivity of [1012] twinning during nanoindentation.

Nanoindentations are performed at temperatures from 25 °C up to 250 °C on the [1010] prismatic surface of a pure Mg crystal. Hertzian contact [56] is employed to establish the relevant stress state at twin onset. We thereby use the pop-in phenomenon to examine the temperature sensitivities of the stresses associated twin formation and growth.

2. Methodology

The as-received material studied in the present work is a rectangular pure Mg single crystal (2 mm in thickness) with a surface parallel to the {1010} plane. This {1010} surface was carefully ground with 1200 grit SiC, followed by 10 min mechanical polishing (MP) of each step using 9 μm, 6 μm, 3 μm water-based diamond suspensions and a 50 nm sized colloidal silica suspension (OPS). Then a subsequent chemical polishing (CP) with an ethanol-based solution (2 ml HNO3 + 1.5 ml HCl + 25 ml ethanol) was performed for 3 min. After chemical polishing, annealing was carried out in a Hysitron TI950 nanoindenter equipped with xSol 600 high temperature stage under argon atmosphere at a flow rate of 2 L/min at 300 °C for 1 hr. Finally, the sample surface prepared for the high temperature tests was chemically polished again for 3 min to remove any oxide layer formed during annealing. The effects of polishing procedure on the pop-in phenomenon is described in Appendix A. After each batch of indentations, the crystal was repolished using the same sample preparation procedures as described above but only from the OPS polishing step. This can effectively “re-set” the sample surface condition for the subsequent testing.

Nanoindentation tests were performed on the annealed and subsequently chemically polished (1010) plane with a spherical tipped conical diamond indenter of a radius in 4.3 μm. The radius of the indenter was verified using a standard contact area analysis on a polycarbonate sample. The Mg single crystal was heated in-situ in the indenter at a rate of 20 °C/s by two resistive heating elements in an environment purged with argon (2 L/min flow rate). Five different indentation temperatures were selected: 25 °C, 100 °C, 200 °C, 230 °C and 250 °C. The load-controlled tests were performed at a constant loading rate of 0.5 mN/s to allow pop-ins to occur at a constant load. Tests were performed to a peak load of 3 mN for 25 °C and 2 mN for 100 °C, 200 °C, 230 °C, and 250 °C (the peak load has no effect on the pop-in events) after preheating the sample for 10 min. 200 independent indentations were carried out for each temperature. And these indentations were spaced at 200 μm intervals. Since we seek to further verify the pop-in deformation mechanism, 10 indent impressions at each testing temperature were measured using the PeakForce error mode on a Bruker Multimode 8 Atomic Force Microscope (AFM) (with resolution up to 0.1 nm) following unloading immediately after the appearance of the first pop-in.
yielding 3. similar J. yielding = is polycrystalline

JMAA 2.

pop-in with chemically loading E at R √ system indentation and the the temperature, E = 39.6 GPa for 250 °C [57]). Although multiple testing shows that the pop-in event occurs in a stochastic manner over a range of loads [45], the four curves in Fig. 2 were selected for their similarity of pop-in load. From the figure, it appears that, for a given pop-in load, an increase in temperature leads to a slightly higher pop-in depth. This result will be confirmed further below.

The indents corresponding to the curves in Fig. 2 were subjected to AFM scanning and the images thus obtained are shown in Fig. 3a. For all temperatures, a twin can be detected by AFM in the indented impression, which is consistent with our contention that a twinning event accompanies the pop-in these tests. Slip lines in Fig. 3a are also observed. These are most likely due to basal slip, as seen in previous studies [53,59]. Interestingly, the slip lines extend to a distance from the indent up to several times impression diameter. The Hertzian stresses far from the indenter are in the order of a Megapascal or less and this is clearly insufficient to nucleate dislocations (which requires the order of a thousand Mega-pascals) so these slip traces must reflect dislocation glide of dislocations nucleated under the indenter. If this activity was due to pre-existing dislocations it would appear at other locations around the indenter on planes that do not intersect with the indenter (see e.g. [52]) but this is not seen here.

To help further rationalize the roles of slip and twinning in the indentation, Fig. 3b shows surface height profiles of corresponding impressions for the 25 °C and 250 °C indentations. The twin morphology is consistent with previous observations [29–31, 58] where identification of the twinning mode with the easily activated {1012} modes was confirmed. In both cases in Fig. 3b, the twin corresponds to a near-linear region on the indent profile section, which is consistent with the fixed shear associated with twinning. The surface displacement accommodated by the twins can be seen to account for between 1/3 and 1/2 of the total displacement. (Some twin shrinkage will invariably have accompanied unloading, e.g. [30,55,60], so these values will under estimate the contribution of twinning to the indent depth and further below we will develop a limiting analysis for the case where the twin spans the entire indent depth.)

The observed twinning displacement in Fig. 3b is clearly too high for the twins to have formed entirely during the small displacement that immediately followed the pop-in event before the test was halted. This affirms the role of twinning in producing the pop-in event. It is also important to note that the indents display curvature and permanent material displacement outside of the twinned region, which belies the presence of plastic deformation in addition to that provided by the twin. Thus significant slip must have accompanied twinning during the pop-in strain excursion.

The load-displacement curves for {1010} indentations at different temperatures taken to a higher equivalent load (2 mN) are shown in Fig. 4a. Only a single well-defined pop-in event was observed in each curve. It is seen that the pop-in load decreases with increasing temperature up to 200 °C for these particular tests but we hasten to add that, due to the stochastic nature of the pop-in, the effect of temperature can

3. Results

Typical load-displacement curves seen during {1010} indentation at room temperature are presented in Fig. 1. For the sample subjected to mechanical polishing, yielding is observed prior to the pop-in, in line with our previous work on polycrystalline alloyed material [28–31]. The curve before yielding follows the well-known Hertzian elastic contact law \( P = \frac{2}{3} E_s \sqrt{R h^3} \), where \( R \) is the radius of indenter tip, \( h \) is the elastic loading depth of indentation at the applied load \( P \), and \( E_s \) is the system reduced modulus.

Fig. 2 shows the load-displacement curves of four tests performed at different temperatures and halted manually after the first pop-in appeared. The elastic responses agree reasonably well with Hertzian predictions (using \( E = 45 \) GPa for room temperature, \( E = 39.6 \) GPa for 250 °C [57]). Although multiple testing shows that the pop-in event occurs in a stochastic manner over a range of loads [45], the four curves in Fig. 2 were selected for their similarity of pop-in load. From the figure, it appears that, for a given pop-in load, an increase in temperature leads to a slightly higher pop-in depth. This result will be confirmed further below.

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Fig. 3. Surface topographies observed after the tests in Fig. 2: (a) AFM images of indented sample surfaces, (b) height profiles of indentation impressions at 25 °C and 250 °C. The red stars and dashed red lines mark the edge of impression. The blue stars and dashed blue lines mark the edge of tensile twin. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 4. (a) Representative load-displacement curves loaded up to 2 mN. (b) AFM images in PeakForce error mode of impressions corresponding to the indentations in (a).

only be ascertained once a significant number of tests have been inspected (see below). After the pop-in Fig. 4a, a region of plastic loading is evident and this is marked by a near linear response in the load-displacement curve with a slope that reduces with temperature. The most likely cause of the slope change is a decrease in the work hardening capacity with temperature. The twins evident in Fig. 4b are notably larger than those in Fig. 3, revealing that stable twin growth accompanies increased penetration of the indenter. Interestingly, the twins are larger in the indents formed at higher temperatures. It appears that both slip and twinning are facilitated at higher temperature. It is likely that this is due to the drop in critical stress for non-basal slip with temperature [9,61], as will be discussed in more detail below.

An examination of the repeat tests reveals that all the tests at room temperature (200 indentations) and 100 °C (200 indentations) showed a well-defined pop-in event. At elevated temperature, not all the tests displayed pop-ins; the tests displaying pop-ins amounted to 196 indentations at 200 °C; 194 indentations at 230 °C and 197 indentations at 250 °C. In the few isolated cases where no pop-in was observed, gradual yielding was seen, and no twin was detected on the surface using AFM scanning. This further validates the proposition that twinning is necessary for a pop-in to appear in the present tests.

The pop-in load distributions obtained for different temperatures are displayed in Fig. 5a. As the temperature rises, the pop-in load distributions move towards lower loads, revealing a marked temperature dependency of the pop-in event. To establish the level of the resolved shear stress on the twinning system corresponding to its activation, the following (Hertzian) expression was employed [33,45]:

\[
\tau_m = \frac{6PE_r^2}{\pi^3R^2}^{1/3}
\]
where $S$ is the ISF defined as the ratio of maximum resolved shear stress on a given slip system to the maximum contact pressure under Hertzian contact, which is 0.306 for tensile twinning [29], 0.18 for basal slip and 0.295 for prismatic slip during [1010] indentation [45]. $E_r$ is the system reduced modulus which only varies slightly with temperature, as evident in Figs. 2 and 4a. The critical stresses on the twinning plane thus calculated are plotted in Fig. 5b. The stresses drop markedly with temperature. This is at odds with the idea that twinning tends to be insensitive to temperature [5] and it likely that the thermal activation of the slip events that accompany twinning are dominating. A rationale is provided in what follows.

4. Discussion

The present study combined nanoindentation with AFM surface examination to correlate tensile twins and pop-in events during indentation of prismatic [1010] surfaces of a Mg single crystal. In every instance where the indent impression was examined (10 indentation impressions for each testing temperature from 25 °C to 250 °C), a single twin was observed. In the few cases where a pop-in did not occur, no twin was observed on the indented surface. This is quite consistent with our previous work on polycrystalline alloyed magnesium using the present experimental geometry [29–31]. Twinning evidently gives rise to the pop-in event in the present experiments and the deformation is elastic up to that point. However, slip lines were also detected on all the surfaces examined, up to a maximum temperature of 250 °C and the deformation is clearly mediated by both slip and twinning.

4.1. Pop-in loads

In the following, we estimate the activation volume and energy associated with the pop-in events and show that these are broadly consistent with what one would expect for the nucleation of lattice dislocations at the surface. Our hypothesis is that the pop-in displacement reflects the rapid expansion of a twin but that the nucleation of the twin requires lattice dislocations to be generated [30]. This is broadly consistent with the simulations of Somekawa et al. [46] which showed that nucleation of basal dislocations during prism plane indentation was not sufficient to provide a pop-in and that further deformation mode activation was required. To verify this argument, we employ the analysis method employed in references [43,44].

Schuh and co-workers [43,44] define an energy barrier $\varepsilon$ required for dislocation nucleation and that the dislocation nucleation rate $\dot{n}$ in a given volume underneath indenter can be described by a reaction rate equation of the standard form:

$$\dot{n} = \eta_0 \exp(-\frac{\varepsilon - \sigma V}{KT})$$

where $\eta_0$ is attempt frequency for dislocation nucleation in a given volume; $\sigma$ is the mechanical stress acting over activation volume $V$; $K$ is the Boltzmann’s constant and $T$ is temperature.

The activation volume can be established via analysis of the stochastic spread of pop-in loads [43]. The energy barrier is then determined from the temperature sensitivity of the loads. The cumulative probability of pop-ins, $F(P)$, as a function of pop-in load, $P$, is given as [43]:

$$\ln[-\ln(1-F(P))] = \alpha P^{1/3} + \xi$$

where $\xi$ is negligibly sensitive to pop-in load and is thus considered to be constant. The term $\alpha$ has the units $N^{-1/3}$ and gives an estimate for the activation volume via [43]:

$$V = \frac{\pi}{S} \left( \frac{3R}{4E_r} \right)^{2/3} KT \alpha$$

where $R$ is the indenter radius (4.3 $\mu$m) and $E_r$ represents the reduced modulus. $K$ is the Boltzmann’s constant. The parameter $S$ is the Indentation Schmid Factor introduced above [29,45]. The parameter $\alpha$ is obtained from the experimental data by plotting them in the form of Eq. (3). This is done in Fig. 6. As in previous work, fitting the equation to the upper part of the curve is obtained [43,44,62,63]. The lower portion of the curve is likely to be highly sensitive to any pre-existing
Table 1
Activation volume calculation.

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>$E_r$(GPa) [57]</th>
<th>$\alpha^b(N^{1/3})$</th>
<th>$V/b^3$.Basal slip dominates ($S = 0.18$)</th>
<th>$V/b^3$.Prismatic slip dominates ($S = 0.295$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>298</td>
<td>45.0</td>
<td>62</td>
<td>2.34</td>
<td>1.43</td>
</tr>
<tr>
<td>373</td>
<td>43.2</td>
<td>98</td>
<td>4.76</td>
<td>2.91</td>
</tr>
<tr>
<td>473</td>
<td>40.8</td>
<td>111</td>
<td>7.11</td>
<td>4.33</td>
</tr>
<tr>
<td>503</td>
<td>40.1</td>
<td>121</td>
<td>8.34</td>
<td>5.08</td>
</tr>
<tr>
<td>523</td>
<td>39.6</td>
<td>166</td>
<td>11.9</td>
<td>7.32</td>
</tr>
</tbody>
</table>

* The value of $\alpha$ is established from the slope in Fig. 6.

Fig. 6. The linear fitting of experimental data plotted in Fig. 5a used to extract activation volume according to Eq. (4). The fitted slopes from the upper part of each curve are 62, 98, 111, 121 and 164 for 25 °C, 100 °C, 200 °C, 230 °C and 250 °C respectively.

defects, which may assist with the onset of plasticity, and is thus omitted from the analysis.

Employing the temperature independent values of $K$ and $S$ (0.18 for basal slip and 0.295 for prismatic slip) in Eq. (4), we obtain the activation volumes shown in Table 1. The values fall in the range of 1.43$b^3$ to 11.9$b^3$ ($b$ is the magnitude of the Burgers vector and equal to $a$ for basal $< a >$ dislocations of Mg, where $a = 0.32$ nm is the Mg lattice constant [64]). Even the large variation of the estimated activation volumes with temperature, this range is broadly consistent with that seen for the nucleation of dislocations at surfaces seen in atomistic modelling (1–10$b^3$ [65]) and lower than what might be expected for the nucleation of a single twin via complex dislocation interactions.

To estimate the energy barrier for incipient plasticity, the approach proposed by Mason et al. [43] is employed. This is based on the following expression [43]:

$$P^{1/3} = \gamma KT + \frac{\pi}{S} \left( \frac{3R}{4E_r} \right)^{2/3} \frac{\varepsilon}{V}$$

Fig. 7. Relation between temperature and $P^{1/3}$ corresponding to the pop-in loads at cumulative fractions 0.7, 0.8 and 0.9. The linear fit is used to determine the $y$-axis intercept, which gives a value of $0.18 \pm 0.005 N^{1/3}$. This is used to estimate the activation enthalpy according to Eq. (6).

where $\gamma$ is assumed to be effectively constant, the energy barrier, $\varepsilon$, can be extracted by plotting $P^{1/3}$ against temperature $T$ and finding the intercept [43]. In Fig. 7, we plot data for the values corresponding to the 70th, 80th and 90th percentile of the cumulative pop-in load distribution. The intercepts fall in the range $0.18 \pm 0.005 N^{1/3}$, which, for the present range of activation volumes, translates to an energy barrier of 0.77 ± 0.34 eV. These values are also broadly consistent with the nucleation of dislocations from a free surface [65]. They are also lower by around an order of magnitude than what might be expected for twin nucleation [1]. It would thus appear reasonable to assume that the nucleation of lattice dislocations triggers the twinning event that gives rise to the pop-in the present material. The mechanism of subsequent twin nucleation is unclear but it may involve the pile-up of dislocations against the interface between the indenter and the sample in a manner analogous to the nucleation of twinning events from internal interfaces [22,66].

4.2. Pop-in depth

It is seen in Fig. 2 that the pop-in depth at constant load is greater at higher temperature. The generality of this ob-
ervation is clear from the plot of pop-in depth against the pop-in load in Fig. 8a. These plots show a strong correlation between pop-in load and depth with an increasing slope with temperature. To analyse this, we consider the limiting case where the pop-in depth is dictated by the extent of the twin ‘burst’ and that the sensitivity to temperature enters predominantly via thermal activation of lattice dislocation activity that relaxes local stresses and which thus enables larger twins to form.

In what follows, this hypothesis is expressed in an analytical form following our previous analysis [30]. The uniqueness of the present approach is that we consider a Hertzian stress field. We seek to show that it yields a reasonable description of the controlling mechanisms. Our analysis is based on estimating the minimum free energy condition that sets the stable twin length and twin aspect ratio in equilibrium with the applied indentation stresses under the indenter. The stability of twin length arises from the fact that the twin propagates away from the indenter and it experiences a reduced driving force generated by the conical indenter. Eventually the net force drops to zero and the twin stabilizes in length. We assume that the relevant driving stress can be approximated by the mean stress, $\overline{\tau}$, sampled by the twin and that this falls off with distance in a manner that can be estimated from the Hertzian stress field. We thereby seek an expression that provides the pop-in depth, $\delta$, as a function of pop-in load, $P$, in a form that contains both an effective twinning ‘friction’ stress required for twin boundary motion and a term that captures the effect of stress relaxation mediated by lattice dislocation activity.

The Gibbs free energy change for an ellipsoidal inclusion is taken as a reasonable approximation for a twin [67]. For the current case of a ‘half’ ellipsoidal shaped twin inclined to the surface, the case is rather complex so we take as a first approximation for the order of the Gibbs energy change a value half that of a full twin. We can thus write [30,67]:

$$\Delta G \sim -\frac{2}{3} \pi l^3 q \left( \overline{\tau} + \frac{\tau_B}{2} - \tau_c \right) + \pi l^2 \varphi$$  \hspace{1cm} (6)

where $q$ is the twin aspect ratio of twin thickness $t$ to twin length $2l$ as described by the half twin, $s$ is the twinning shear, $\tau_B$ is the misfit back stress [13,68,69] and $\overline{\tau}$, $\tau_c$, $\varphi$ are the mean applied shear stress, friction stress for twin growth and twin surface energy, respectively. For an elliptical twin in magnesium with length twice its width (roughly the shape of the present twins [30]), the Eshelby misfit back stress can be estimated as $\tau_B \sim 2.7sqG/\beta$, where $G$ is the shear modulus and $\beta$ is a relaxation coefficient introduced to capture the influence of plastic relaxation effects (lattice dislocation nucleation and/or glide) occurring in the vicinity of the twin [30,70]. The numerical role of $\beta$ is to reduce the magnitude of the misfit back stress generated in the twin as a result of slip activity in the surrounding matrix. A large value of $\beta$ indicates a high level of nearby slip activity and correspondingly lowered twin misfit stress. Since the scale of a twin during a pop-in event in the present work extends to the micron level, the volume term dominates and for a first approximation can safely be obtained neglecting the surface energy term in Equation 6.

Differentiating Eq. (6) with respect to the twin back stress and setting to zero enables identification of the minimum energy condition, which arises when $-\tau_B = \overline{\tau} - \tau_c$. The free energy change at this condition is:

$$\Delta G \sim -\frac{\pi}{8.1} \frac{\beta l^3}{G} \left( \overline{\tau}^2 - 2\overline{\tau} \tau_c + \tau_c^2 \right)$$  \hspace{1cm} (7)

We seek to establish the subsequent minimum of this expression as the twin increases in length and experiences a decreasing mean stress as it propagates out of the stress concentration.

Mean stresses, $\overline{\tau}$, were calculated from Hertzian stress fields for simulated twins with an aspect ratio, $q$, from 0.04 to 0.08 placed immediately beneath the indenter but intersecting the surface at the edge of the contact circle and habiting a plane inclined approximately 45° to the indenter surface [30]. The results are shown in Fig. 8b and it can be seen that the
mean stress can be approximated by a power law:

$$\tau = k l^{-n}$$  \(8\)

where \(k\) is an adjustable parameter that gives the mean stress for twin length, \(l\), under the contact radius, \(a'\) (valid for \(l > a'\)). The values of \(k\) and \(n\) will depend upon the twin shape, twin plane and the nature of the stress field. Fig. 8b shows that Eq. (8) provides a reasonable approximation for \(k = \frac{2}{3}\) and \(n = \frac{2}{3}\).

Substituting Eq. (8) into Eq. (7) to eliminate \(l\) and differentiating with respect to the mean twin stress reveals that the minimum condition arises when:

$$\tau = \tau_c \left( \frac{3}{3 - 2n} \right)$$  \(9\)

For \(n = \frac{1}{3}\), we see that the twin will extend until the mean stress drops to a value of \(2\tau_c\). (Interestingly, the value obtained in our previous analysis - \(9\tau_c / 5\) - where the stress field was approximated using plasticity analysis is similar in size [30]). We can now write expressions for the equilibrium twin aspect ratio and twin length:

$$q = \frac{l}{2l} = \beta \frac{\tau - \tau_c}{2.73G} = \beta \frac{\tau_c}{2.73G}$$  \(10\)

$$l = a' \left( \frac{a_m}{\tau} \right)^{1/n} = a' \left( \frac{1}{\tau_c} \right)^{1/n}$$  \(11\)

The Hertzian contact radius, \(a'\) prior to the pop-in can be given by \(a' = (3PR/4E)^{1/3}\) [45,56] and the resolved maximum Hertzian shear stress, \(\tau_m\), is given by Eq. (1). We can also note that if the pop-in depth, \(\delta\), is dominated by the twinning shear, we can approximate it as \(\delta = t s \sqrt{\frac{\lambda}{\pi}}\) [28]. Therefore, the twin growth stress can be written as:

$$\tau_c \approx \frac{2}{5} \beta^3 \left( \frac{S}{\pi} \right)^4 \left( \frac{1}{G} \right)^{3} \left( \frac{P^{*}}{R} \right)^{5/3} \frac{\left( \frac{P^{*}}{R} \right)^{2}}{\delta}$$  \(12\)

This expression suggests that the pop-in depth should vary with \(P^{*}\) and Fig. 8a shows that this is born out more-or-less by the experiments (if we allow for a small degree of displacement - 300 \(\mu\)N - from the origin due to instrument error). Fitting \(P^{*}\) = \(\lambda \delta + c\) to the data in Fig. 8a (where \(\lambda\) is a coefficient containing all the other terms of Eq. (12) and \(c\) is an instrument constant) enables Eq. (12) to be plotted for the different temperatures employed in the present study. This is done in Fig. 9 using the parameters in Table 2.

![Fig. 9. The relationship between twinning friction stress and the relaxation factor \(\beta\) for different indentation temperatures - estimated by fitting Eq. (12) to the data in Fig. 8 (see Table 2).](image)

![Table 2](image)

Table 2

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>(\lambda / 10^4) (N/m)</th>
<th>(E) (GPa)</th>
<th>(G) (GPa)</th>
<th>(R (\mu m))</th>
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<td>45.0</td>
<td>17.0</td>
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</tr>
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<td>43.2</td>
<td>15.9</td>
<td>4.3</td>
</tr>
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<td>40.8</td>
<td>15.4</td>
<td>4.3</td>
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<td>3.2</td>
<td>39.6</td>
<td>14.9</td>
<td>4.3</td>
</tr>
</tbody>
</table>

\(\tau\) is the fitting coefficient employed to plot the lines in Fig. 8a.
in magnesium alloys [9,61]. The use of a single relaxation factor to capture a phenomenon as complex as the slip occurring in the vicinity of a twin will be problematic. The use of single crystals in the present work also introduces a significant effect because of the absence of the relaxation mechanisms available at grain boundaries. Nevertheless, the present data and our analysis show that it is almost impossible to disentangle twinning from concurrent slip events in a magnesium. The apparent temperature sensitivity of the twinning events in nanoindentation is likely to be due to the temperature sensitivity of dislocation glide.

Finally, a comment is in order on how the present findings might relate to the behaviour of bulk polycrystalline material. In such material, there is likely to be a sufficient dislocation density present to enable dislocation mediated twin nucleation without the need to nucleate dislocations. One would thus not expect to see the temperature sensitivity of twin nucleation we have detected in the present nanoindentation experiments. The enhanced plastic relaxation that we propose to be responsible for larger pop-ins at elevated temperature is something expected to be seen in polycrystalline material. As noted above, the result tallies well, qualitatively, with observations of thicker twins at elevated temperature [13]. Evidently, in interpreting the temperature sensitivity of twinning, the temperature sensitivity of any associated lattice dislocation activity should be considered.

Conclusions

A near defect free {10\bar{1}0} surface on an Mg single crystal was prepared by surface polishing and annealing. This was used to investigate the temperature dependency of pop-in events that marked the elastoplastic transition during nanoindentation. The following are the conclusions:

- Single {10\bar{1}2} twins can be identified in AFM images of the surface just after the pop-in for each temperature examined up to 250 °C. For indentation without a pop-in, no twin was observed after unloading. We thus confirm our previous findings that for nanoindentation of {10\bar{1}0} surfaces in magnesium, the pop-in arises from a twinning event.
- Slip is seen to accompany the pop-in event as evidenced by basal slip lines observed on the sample surface and the residual curvature of the indent impression.
- With increasing temperatures, the load corresponding to the first pop-in dropped and the pop-in depth slightly increased (for an equivalent load).
- Thermal activation analysis revealed that the temperature dependence of the pop-in load is broadly consistent with dislocation nucleation at the sample surface. It is thus proposed that dislocation nucleation triggers the twinning event that creates the pop-in the present tests.
- The small temperature dependence of the depth of the pop-in can be understood to arise from the temperature dependency of the lattice dislocation mediated plastic relaxation that accompanies twinning.

Declaration of Competing Interest

None

Acknowledgements

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Appendix A

In order to acquire consistent sample surface condition for high temperature testing, systematic annealing experiments

![Fig. A1. Room temperature pop-in load statistics for different sample preparations using mechanical polishing (MP), chemical polishing (CP) and annealing in argon. (a) Tests performed sequentially on the one sample. Red and black symbols refer to the first and second series of tests respectively. (b) Room temperature pop-in load statistics for samples subjected to mechanical polishing (MP), chemical polishing (CP) and annealing in argon at 250 °C.](image-url)
were performed at 250 °C and 300 °C in the nanoindenter chamber. Room temperature indentations on successively annealed samples were carried out to determine the response. Fig. A1a shows the data plotted as cumulative fraction of tests against pop-in load for samples produced by different polishing techniques. The red symbols in Fig. A1a show how the pop-in load obtained by indenting the as-annealed surface is influenced by annealing time at 300 °C after chemical polishing. These annealing treatments lower the pop-in load. Our hypothesis is that this is due to the formation of a thin oxide film [73,74]. Lowering the annealing temperature to 250 °C resulted in no change in pop-in load and so this set was as the maximum temperature for the present nanoindentation tests (Fig. A1b).

When the samples annealed 4 h at 300 °C are subjected to further mechanical and chemical polishing (10 min OPS polishing following by 3 min chemical polishing), the pop-in load distribution returns to the pre-annealed levels, showing the present samples can be ‘reset’ following annealing (black solid circles in Fig. A1b).

To obtain surfaces with minimum defect content, annealing was followed by chemical polishing and the effect of annealing time was examined (red symbols in Fig. A1a). After these treatments, consistent pop-in distributions are obtained. These display pop-in loads higher than all other treatments, which attests to a low surface defect content. Based on these findings, the surface preparation employed for the present study involves: mechanical polishing (MP) + OPS polishing for 10 min + chemical polishing (CP) for 3 min + 300 °C annealing for 1 h + chemical polishing for 3 min.

References