The effect of crystal orientation on the indentation response of commercially pure titanium: experiments and simulations

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This study combines nanoindentation, electron backscatter diffraction (EBSD) and crystal plasticity finite element analysis to examine the anisotropy in the indentation behaviour of individual grains within an α-Ti polycrystal. Nanoindentation is utilized to mechanically probe small volumes of material within grains for which orientations are known from prior EBSD mapping. Both indentation modulus and hardness decrease significantly as the indentation axis is inclined further from the c-axis; the plastic response showing the more marked anisotropy. Recently developed high angular resolution EBSD has been utilized to examine selected indents, providing maps of elastic strain variations and lattice rotations. From such maps lower bound solutions for the density of geometrically necessary dislocations (GNDs) have been established. Crystal plasticity modelling showed promise in capturing correctly the orientation dependence of load–displacement response and in lattice rotations local to the indenter, particularly for indentation into a basal plane which generated threelfold rotational symmetry about an axis parallel with the indentation direction which was also observed in experiments.

Keywords: electron backscatter diffraction; nanoindentation; finite element modelling; crystal plasticity

1. Introduction

Improving the performance of novel titanium alloys for use in extreme conditions in aerospace and automotive applications relies upon controlling the themomechanical processing to create a suitably textured microstructure. To understand how the texture affects the mechanical properties, one must first examine mechanical properties of single crystals and interpret the deformation processes involved. Once the single crystal and grain boundary behaviours have been understood, processing methods aimed at generating desired micro-textures can be developed and utilized (Rugg et al. 2007).

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The mechanical properties of single-crystal α-Ti are highly anisotropic (Jaffee et al. 1970), yet few studies have been able to test a wide variety of crystal orientations. Many different single crystals are required to gain a comprehensive understanding of the effect of orientation on mechanical properties. The cost and difficulty of producing large single crystals suitable for macroscale mechanical testing prohibit extensive testing programmes. If smaller scale tests are employed, one could use a large-grained polycrystalline material to test a wide variety of orientations.

Nanoindentation is an instrumented hardness testing technique, with precise indent location, high-resolution load control and displacement measurement (Oliver & Pharr 2004). During indentation one can measure continuous load–displacement, and after indentation the modulus and hardness results can be calculated from the unloading portion of the experiment (Pharr & Bolshakov 2002).

Nanoindentation probes a very small volume around the indenter and when combined with a fully characterized large grained polycrystalline material, a wide variety of crystal orientations can be probed, which provides information on the materials performance on the nano- and micro-scales. Increasingly, these tests are being used to improve our understanding of materials performance on these scales.

It is readily apparent that a range of different length scales are typically found in titanium alloys, where the behaviour of individual laths, colonies of laths and prior beta grains has been shown to influence the macroscopic mechanical properties (Rugg et al. 2007).

Commercially pure (CP) titanium is widely used, typically in highly oxidizing and corrosive environments in the aerospace industry (Boyer 1996), in chemical and process engineering (Yamada 1996), and in a variety of biomedical applications (Geetha et al. 2009); large grain CP Ti is also an excellent model system to examine deformation in alpha grains of other Ti alloys.

Viswanathan et al. (2005) used transmission electron microscopy (TEM) to directly examine the dislocation structures around indents made in different grains of Ti64. In the study they noted an increase in hardness near the [0001] indentation axis and attributed the hardening to a high density of \( \langle c + a \rangle \) type geometrically necessary dislocations (GNDs) directly below the indentation; the softer grains had limited \( \langle c + a \rangle \) type GNDs and many more \( \langle a \rangle \) type GNDs. The authors did not comment on any variation in modulus with orientation, as the contribution of surrounding grains would distort any effect with such a small grain size (20 μm). Merson et al. (2008) have recently studied the effect of crystal orientation on the indentation hardness of CP titanium, reporting similar trends as Viswanathan et al. (2005).

Others have examined the elastic and plastic zones of nanoindents utilizing conventional electron backscatter diffraction (EBSD). The EBSD technique allows larger, more realistic and more deformed samples to be characterized compared to TEM-based studies (Demir et al. 2009; Rester et al. 2008; Zaafarani et al. 2008).

By utilizing focused ion beam–scanning electron microscope (FIB–SEM) tomography equipped with EBSD, Demir et al. (2009) were able to examine the subsurface GND densities below nanoindents made in copper. They attempted to quantify the indentation size effect (ISE) in relation to the
GND content and observed that there was a heterogeneous GND density surrounding the indents, in contradiction to the majority of existing theories on the ISE.

While FIB–SEM and EBSD tomography enables all nine components of the lattice curvature to be obtained there may be relaxation issues related to the exposure of a fresh surface at each milling stage. This is especially true with areas of high dislocation density, such as those found directly under the indent. Also great care is required to correctly align (translation and rotation) EBSD data taken after each FIB milling stage.

Conventional EBSD analysis utilizes the Hough transform to locate and index the bands present in a Kikuchi pattern. The bands identified are compared to a look-up table based on the underlying crystallography of the specimen, and a matrix describing the rotation of a reference frame to the crystal orientation can be calculated (Schwartz et al. 2000). Hough-based EBSD analysis is limited to an angular resolution of ±0.01 to 0.02 rad and results in uncertainty in the axis of misorientation between two points (Humphreys 2001) which can be very large when the misorientation angle is small (Wilkinson 2001). (These uncertainties reduce the accuracy of any lattice curvature measurement and inhibit resolving dislocation structures reliably onto specific slip systems.)

To avoid the pitfalls of Hough-based EBSD analysis, Wilkinson et al. (2006a) used cross-correlation to map small distortions in patterns from a strained crystal compared to an undistorted reference pattern from strain-free material. This method has improved the measurement of lattice rotations to an angular sensitivity of ±0.0001 rad, and enables specific misorientation axes to be defined (elastic strains are also determined at a sensitivity of $1 \times 10^{-4}$). They utilized the same capture angle as conventional EBSD but captured patterns at the full resolution (1 × 1 binning) and bit depth (12 bit) on a peltier-cooled CCD camera as the scan progressed. This technique has been used to examine elastic strain in semiconductor materials (Wilkinson 2006), the lattice rotations associated with a fatigue crack in a nickel alloy (Wilkinson et al. 2006b), and elastic strains near wedge indents in Si (Vaudin et al. 2008).

### 2. Simulating crystal plasticity during indentation in Ti alloys

In this section, we introduce the crystal plasticity finite element modelling techniques employed later to simulate indentations into CP Ti single crystals. The model takes account of the basal, prismatic and pyramidal slip systems, elastic anisotropy, and the (moderate) rate sensitivity demonstrated by CP Ti, even at room temperature.

The overall plastic velocity gradient is made up of contributions from all active slip systems and is given in terms of the slip directions $s^\alpha$ and slip plane normals $n^\alpha$ corresponding to the $\alpha$th slip system by

$$ L^p = \sum_{\alpha=1}^{n} \dot{\gamma}^\alpha s^\alpha \otimes n^\alpha $$

and the slip rate $\dot{\gamma}^\alpha$ is taken from the model developed by Dunne et al. (2007)

$$\dot{\gamma}^\alpha = \rho_{\text{SSD}}^m b^2 \nu \exp \left( \frac{\Delta F}{kT} \right) \sinh \left( \frac{(\tau^\alpha - \tau_c)\Delta V}{kT} \right),$$

(2.2)
in which $\rho_{\text{SSD}}^m$ is the density of mobile statistically stored dislocations, $\Delta F$ is the Helmholtz free energy barrier which needs to be exceeded in order for a pinned statistically stored dislocation to be released, $b$ is the magnitude of the Burger’s vector, $\nu$ is the frequency of dislocation jumps (successful or otherwise), $k$ is the Boltzman constant, $T$ is the absolute temperature. $\tau^\alpha$ is the resolved shear stress, $\tau_c$ is the critical resolved shear stress (CRSS), and $\Delta V$ the activation volume associated with dislocation climb, which is taken to be $\Delta V = \lambda b^2$, where $\lambda$ is the pinning distance. Here it is assumed that the predominant pinning mechanism is that caused by the development of GNDs and immobile statistically stored dislocations (SSDs). The pinning distance is therefore related to the density of GNDs $\rho_{\text{GND}}$ and the density of immobile SSDs $\rho_{\text{SSD}}^{\text{im}}$ and in the absence of more detailed information, is assumed to be given by

$$\lambda = \frac{1}{\sqrt{\rho_{\text{SSD}}^{\text{im}} + \rho_{\text{GND}}}}.$$  

(2.3)

The densities of mobile and immobile SSDs are taken to be the same and initially assigned the value $1 \times 10^{10}$ m$^{-2}$. It is the case that the density of SSDs increases during subsequent deformation but at what rate is unknown in this material under conditions of indentation. The incorporation of the evolution of SSD density would currently require the inclusion of an empirical evolution equation into the crystal model, though eventually suitable experimental data and understanding may become available to allow this to be done in a more physically argued way. Thus for reasons of simplicity, and retaining a physical basis, we choose not to evolve the SSD density directly with subsequent deformation, but do allow density to change as a result of the GND evolution which is described below. Cognizance of this simplification is necessary when interpreting the model predictions.

The density of GNDs is obtained from the relation between inelastic strain gradients and dislocation densities originally proposed by Nye (1953) and subsequently developed by Busso et al. (2000) to define a tensorial measure of GND density related to the resultant Burger’s vector of the GNDs.

In the present model, in the absence of other information, we require the magnitude only of the density of GNDs for the slip rate coupling given in equation (2.5), and this is obtained from Liang et al. (in preparation) as

$$\rho_{\text{GND}} = \frac{1}{b_{\text{GND}}} |\text{curl}(F^p)|.$$  

(2.4)

Equation (2.4) is geometrically determined; it contains no material properties and the density of GNDs is obtained purely from knowledge of the plastic deformation gradients. The absence of a plastic strain gradient requires a density of GNDs of zero. The GND density is related to non-uniform plastic deformation and to the curvature of the crystal lattice; it is that portion of the total density specifically needed to maintain the continuity of the crystal lattice. If, however, with progressive deformation, the local plastic strain gradient diminishes, the

consequence of equation (2.4) is that the density of GNDs also correspondingly decreases. What happens in reality remains an open question, but it is unlikely that the overall density of dislocations diminishes during deformation. We can suppose that GNDs are no longer necessary to support a plastic strain gradient and therefore are converted to SSDs. If the GND density drops by $\Delta \rho_{\text{GND}}$, then two extreme possibilities are that: (i) additional dislocations are introduced to reduce the curvature previously generated and that these are stored as SSDs along with a matching contribution converted from GNDs; (ii) again additional dislocations are generated but instead of being stored in the material they react with some existing GNDs and both are annihilated. Situation (i) leads to SSD density increasing by $2 \Delta \rho_{\text{GND}}$ while for (ii) the SSD density remains unchanged. Our model is intermediate between these two cases and is described by

$$\text{If } \rho_{\text{GND}}^{t+\Delta t} < \rho_{\text{GND}}^t, \text{ then } \rho_{\text{SSD}}^{t+\Delta t} = \rho_{\text{SSD}}^t + (\rho_{\text{GND}}^t - \rho_{\text{GND}}^{t+\Delta t}).$$

The slip rule and dislocation model are coupled in the large deformation crystal plasticity non-local formulation and implemented within the finite element code ABAQUS. A user defined element (UEL) was developed in order to determine the plastic strain gradients, as described elsewhere by Dunne et al. (2007), and a detailed description of the large deformation formulation may be found in Liang (2008).

The Berkovich indenter and the single-crystal titanium sample are shown schematically in figure 1. In the simulations the titanium sample is modelled as a hexagonal close packed (HCP) single crystal with dimensions of $20 \mu m \times 20 \mu m \times 5 \mu m$. We assume a rigid indenter penetrating a homogeneous anisotropic single crystal. Contact between the indenter and the sample was assumed to be frictionless as previous indentation simulations by Wang et al. (2004) showed no significant influence of friction on the analysis.

A three-dimensional finite element mesh with 9000 20-node brick elements with reduced integration is used for the discretization of the sample as shown in figure 1. The lower surface of the sample is fixed in all directions. The side and top surfaces are free to deform in any direction. The mesh is finer close to the indenter tip to allow for good resolution of the contact area.

Figure 1. Finite element mesh composed of 9000 20-noded reduced integration three-dimensional elements, used to model the single-crystal pure titanium sample.
While fixed in three-dimensional space, it is useful to bear in mind that the finite element sample modelled is smaller than that used in the experiment (for reasons of computational time) and the question arises, therefore, about the most appropriate boundary conditions to apply to capture as accurately as possible the experimental conditions. It is likely that the appropriate boundary conditions lie somewhere between the two extremes of fixing the free sides in all directions or allowing complete freedom of displacement. In order to determine the most appropriate boundary conditions, we carried out simulations of indentation to 1000 nm with both sets of boundary conditions given above, with the overall result that the differences in load-indentation depth and in lattice rotations resulting from the differing boundary conditions were small. In addition, while not significantly different, we obtained a better representation of the experimental load–displacement behaviour with boundary conditions in which the free side surfaces were not constrained and, for this reason, these boundary conditions have been retained for all subsequent analyses. (Figures showing the differences in boundary conditions can be found in the electronic supplementary material.)

The indenter modelled is given a tip radius of 1000 nm, which is larger than that in the experiment. However, previous work by Liang et al. (2008) has shown that the results obtained from the somewhat ‘blunter’ indenter converge with those obtained with the accurate indenter geometry for indentation depths greater than about 300 nm. The primary advantage of the blunter indenter geometry is the very significant reduction in computational time which results.

The (anisotropic) elastic constants used for titanium are as follows: $C_{11} = 162.4$ GPa, $C_{33} = 180.7$ GPa, $C_{14} = 117$ GPa, $C_{66} = 35.2$ GPa, $C_{12} = 92.0$ GPa and $C_{13} = 69.0$ GPa, as noted by Viswanathan et al. (2005). The other material properties required are: Burger’s vector for $\langle a \rangle$ type slip $b = 2.95 \times 10^{-10}$ m, and for $\langle c + a \rangle$ type $b = 5.53 \times 10^{-10}$ m, Boltzman’s constant $k = 1.381 \times 10^{-23}$ JK$^{-1}$, the frequency of dislocation jumps $v = 0.79 \times 10^{13}$ s$^{-1}$ from Cottrell (1995), and the activation energy $\Delta F = 1.54 \times 10^{-19}$ J is chosen to ensure there is no significant strain rate effect. The CRSSs were chosen to ensure the computed load–displacement curves closely match those obtained in the experiments. The value used for $\langle a \rangle$ type slip systems is 210 MPa and that for $c + a$ type slip systems is four times the value for $\langle a \rangle$ type slip according to Viswanathan et al. (2005), giving 840 MPa. The initial density of statistically stored dislocations is taken to be $\rho_{SSD} = 5 \times 10^{10}$ m$^{-2}$, and the initial density of GNDs is taken to be zero.

3. Experimental method

A bar of grade 1 CP titanium (700 ppm O) was obtained from Timet UK and heat treated at 830°C for 24 h to create an average grain size of 130 μm. A 2 mm deep sample of area 10 × 10 mm was cut from the bar and mechanically ground to 2500 grit. It was then repeatedly polished using a mixture of 0.04 μm colloidal silica and hydrogen peroxide (5 : 1) and etched using 1 per cent HF, 10 per cent H$_2$NO$_3$ in water until crisp polarized light images showing the grain structure clearly could be obtained. This typically takes four repeated steps of the polish and etch procedure.

A 15 × 16 array of nanoindents with 70 μm spacing was made using a NanoindenterXP (MTS Instruments) equipped with a diamond Berkovich tip (tip radius approx. 40 nm). A constant target indentation strain rate of 0.05 s⁻¹ was used to a target peak depth of 1 μm. At the peak depth, the indenter was held at the maximum resultant load for 15 s to allow for any creep and then unloaded at a rate of 0.1 mN s⁻¹ to 10 per cent of the peak load. The indenter was held at this load for a further 50 s to correct for any thermal drift. Unloading hardness and modulus values were calculated from the first 50 per cent of the resultant unloading curves using Testworks 4 software (MTS Instruments).

Topographical atomic force microscopy (AFM) was conducted in contact mode on a Pacific Nanotechnology Nano-R with SPM Cockpit 2002. Images were analysed with Gwyddion 2.4. EBSD was conducted in a JEOL-6500FEG SEM with TSL OIM DC v4 software, at 20 kV and using a probe current of approximately 1 nA. Orientations for each indented grain were extracted from a low resolution, large area map (∼ 1 × 1 mm) of the indented region. Maps (30 × 30 μm) of selected indents were made with a step size of 0.3 μm. These maps were for high-resolution EBSD analysis and patterns were captured at 1 × 1 binning and recorded to disk for subsequent analysis, for elastic strain and lattice rotations, using CrossCourt v2 (BLG Productions). CrossCourt produces a pair of ‘data quality’ parameters which can be used to remove data that may have a large error, due to excessive pattern distortion and blurring or shadowing from surface topography such as those that occur inside the indent. To reduce the effect of pattern noise further, the reference pattern was averaged from 25 frames taken far away from the indent, but within the same grain. Subsequent data analysis for GND distributions and data visualization was undertaken using routines developed within Matlab. The custom Matlab software first removes data with either a normalized average cross-correlation peak height less than 0.4 or a mean angular error greater than 40 mrad, corresponding to poor data.

4. EBSD analysis of GND content

Distortions to the crystal cause small shifts in the positions of the zone axis, as well as other features, in the EBSD patterns (EBSPs). Shifts can be measured at four or more regions of interest (ROI) by two-dimensional cross-correlation between reference and distorted EBSPs. Each ROI is transferred into the Fourier domain, for fast cross-correlation. Band pass filtering of the ROI can be performed while in the Fourier domain, so as to reduce the effect of pattern noise on the measured shifts. These shifts can be used to calculate the elastic displacement gradient tensor, \( \beta \), which is often denoted by \( F^e \) in the mechanics literature:

\[
\beta = \begin{pmatrix}
\frac{\partial u_1}{\partial x_1} & \frac{\partial u_1}{\partial x_2} & \frac{\partial u_1}{\partial x_3} \\
\frac{\partial u_2}{\partial x_1} & \frac{\partial u_2}{\partial x_2} & \frac{\partial u_2}{\partial x_3} \\
\frac{\partial u_3}{\partial x_1} & \frac{\partial u_3}{\partial x_2} & \frac{\partial u_3}{\partial x_3}
\end{pmatrix}
\] (4.1)
Note that it is impossible to separate the \((\partial u_i/\partial x_i)\) components as the final degree of freedom, the hydrostatic dilation or contraction of the lattice does not result in a change in angles between the zone axes.

\(\beta\) can be calculated by measuring the shifts at four or more ROI across the pattern and, like Wilkinson et al. (2006a), we achieved a more accurate solution by using 20 ROI and thus solving for an over-determined set of equations by a least-squares method. Wilkinson (2006) reports a standard deviation of 0.1 mrad for rotations and \(10^{-4}\) in strain.

Assuming a plane stress condition \((\sigma_{33} = 0)\) to prevail within the few tens of nanometres, from which depth EBSPs are generated, the terms on the leading diagonal of the deformation tensor \((\partial u_i/\partial x_i)\) can be separated.

Finally, the elastic displacement gradient tensor is transformed to the sample frame and the following can be calculated (assuming small displacements/strains):

1. Elastic normal strains, \(\varepsilon_i = \frac{\partial u_i}{\partial x_i}\); (4.2)
2. Elastic shear strains, \(\varepsilon_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)\); (4.3)
3. Lattice rotations, \(\omega_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} - \frac{\partial u_j}{\partial x_i} \right)\). (4.4)

Several authors (Adams 1997; Sun et al. 1998; El-Dasher et al. 2003; Field et al. 2005; Kysar et al. 2007; Larson et al. 2007; Demir et al. 2009; Pantleon et al. 2008; Zisman et al. 2008; Miyamoto et al. 2009) have used the lattice curvatures to calculate densities of GNDs within samples, using Hough-based EBSD, which has a lower angular resolution and thus is severely limited for GND analysis (the lower bound is capped by angular noise, at approx. \(1 \times 10^{14}\) GNDs m\(^{-2}\)). Using high angular resolution EBSD, six of the nine lattice curvature components can be calculated from the changes in lattice rotation across the sample surface \((\partial \omega_{ij}/\partial x_k)\); the remaining three concern variation of the lattice rotations with depth below the surface. Using these six components it is possible to calculate a lower bound solution to the density of GNDs using Nye’s (1953) dislocation analysis. However, one must be careful as elastic strain gradients could also contribute to the lattice curvatures, and unfortunately the gradients along the depth direction of different strain components (i.e. \(\partial \varepsilon_{ij}/\partial x_3\)) contribute to six of the nine terms in the distortion tensor. If the elastic strain gradients measured are sufficiently small, compared to the rotation gradients, it would be unlikely that those which cannot be measured are sufficiently large as to contribute to the curvatures significantly. Furthermore, in situations where the rotation gradients dominate, reducing the curvatures to just three would be unjustified given that the vastly reduced constraints on the dislocation content tends to a markedly lower lower bound solution. Therefore, we take the approach that if we check that it is reasonable to neglect the elastic strain gradients then we can continue with the analysis just using the six measured rotation gradients.

The lattice curvatures are linked to the GND densities by solving equation (4.5) for \(\rho\):

\[
\mathbf{K} = (\mathbf{A}_1, \mathbf{A}_2, \mathbf{A}_3 \ldots \mathbf{A}_S)(\rho),
\] (4.5)
where the column vector $\mathbf{K}$ is populated with the six curvature components and $\mathbf{A}_s$ is populated from the tensor product of the Burger’s vector and line direction of the $s$th dislocation type (i.e. $\mathbf{A}_s = \mathbf{b}_s \otimes \mathbf{l}_s$).

In $\alpha$-Ti the following slip systems are known to be possible and have been used in defining the $\mathbf{A}$ matrix, by using their Burger’s vectors and line directions: $\langle c + a \rangle$ screw (6); $\langle c + a \rangle$ edge on pyramidal slip planes (12); $\langle a \rangle$ screw (3); $\langle a \rangle$ edge on basal slip planes (3); $\langle a \rangle$ edge on pyramidal slip planes (6); and $\langle a \rangle$ edge on prismatic slip planes (3), see Jones & Hutchinson (1981). The $\langle c + a \rangle$ type dislocations are generally required for this analysis as $\langle a \rangle$ type dislocations are insufficient to support all measured curvatures (the rank of $\mathbf{A}$ is 5 with only $\langle a \rangle$ type dislocations, and is 6 when $\langle c + a \rangle$ type are considered).

As equation (4.5) is under-determined, with potentially 33 unknown GND densities described by only six curvatures, we have used a standard linear programming algorithm (in Matlab) to find a lower bound solution of the GND densities for each individual slip system, at each point in the EBSD maps.

In order to produce a stable solution, we ‘unfold’ the dislocation types to consider positive and negative types separately (by considering oppositely signed Burger’s vectors in constructing $\mathbf{A}_s$). This results in $33 \times 2$ slip systems in total. We can now constrain each GND density to be zero or greater. Finally, we choose a solution, which may or may not be unique but does support the lattice curvatures, and also generates the lowest possible total GND line energy. For the different GND types the line energies are proportional to $b^2$ for screw dislocations and $(b^2/(1-\nu))$ for edge dislocations (where $\nu$ is Poisson’s ratio). This weights the use of the GND types in supporting the curvatures as follows (lower is more favourable): $\langle a \rangle$ screw $-0.087$, $\langle a \rangle$ edge $-0.124$; $\langle c + a \rangle$ screw $-0.306$; $\langle c + a \rangle$ edge $-0.437$.

Finally, we can fold each positive and negative pair of dislocation types back together and then calculate the total GND density by summing up (the absolute values of) all the GND densities for each slip system used.

5. Experimental results

(a) Effect of orientation on mechanical properties

Of the 240 indents in the array only 147 indents are used to compare the variation of mechanical properties with orientation, as the remaining indents either were not successfully made or were within approximately 20 $\mu$m of a grain boundary, resulting in the elastic and plastic zones extending into two or more crystals.

In order to quantify the effect of orientation on mechanical properties, only the variation in declination angle (the angle between the crystal’s $c$-axis and the indent loading direction) has been considered.

Figure 2 shows that both the hardness and modulus are highest when indenting into a basal plane ($c$-axis parallel to the loading direction) and lowest when indenting into a prismatic plane ($c$-axis perpendicular to the loading direction). The anisotropy is slightly greater in the plastic response (hardness) than in the elastic response (indent modulus) though both are significant at 15–20% of the mean.
Figure 2. Variations in the unloading modulus and hardness with declination angle. Filled grey circles, unloading modules; filled black triangles, unloading hardness.

(b) High-resolution EBSD analysis of two indents

Two indents were selected from the array, one with loading direction nearly parallel to the c-axis (indent a), the second nearly perpendicular (indent b), which corresponded to the following Bunge Euler angles (phil, PHI, phi2): \( a = (321°, 9°, 240°) \), \( b = (176°, 88°, 39°) \), as shown schematically in figure 3.

Elastic hoop, radial and in-plane shear strains referred to an origin at the centre of the indent are shown in figure 4 (the vertical striation seen on maps of indent a was caused by contamination in the SEM).

There is considerable pile-up near to the top and bottom corners of indent b. The extensive deformation and change in surface plane due to the topography leads to poor quality EBSPs. Data in these regions are not shown because results from the cross-correlation analysis fall below the two ‘data quality’ thresholds described in §3.

The lattice rotations shown in figure 5 show the size and shape of the plastic zone associated with each indent. Neither plastic zone is homogeneous with respect to the indenter; this is especially evident in the in plane rotations associated with indent a, where there is a threefold rotational axis about the \( \langle c \rangle \) direction, resulting in three pairs of positive and negative lobes, with mid-lines parallel to the \( \langle a \rangle \) directions. The in-plane rotations for indent b have similar symmetry about the \( \langle c \rangle \) direction.

Lattice rotations are slightly larger than elastic strains and are markedly more localized around the indenter. The variation of elastic strains is generally smoother and over larger distances.

The three lattice rotation plots have been numerically differentiated in the \( x_1 \) and \( x_2 \) directions, to produce six of the nine lattice curvature plots, and are presented in the electronic supplementary material figures.
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From these curvatures, the custom Matlab software calculates the lower bound GND solution, on each individual slip system. The magnitudes of these solutions can be summed to produce densities based upon the slip systems mentioned in the introduction, with deconstructions showing only \( \langle a \rangle \) type; only \( \langle c + a \rangle \) type; and total HCP GND densities (figure 6). For both indents, the density of \( \langle a \rangle \) type is approximately 10 times greater than the density of \( \langle c + a \rangle \) type dislocations and the GND density plots for both indents show symmetry around their \( \langle c \rangle \) axis.

Plots showing the deconstruction onto individual slip systems, used to construct the total GND density, can be found in the electronic supplementary material figures. It should be emphasized that this presents a set of GNDs that support the measured lattice curvatures while minimizing the dislocation line energy, there are many other combinations of GNDs that can generate the same lattice curvatures.

(c) Atomic force microscopy of two indents

AFM was performed on two indents of similar orientations to indent \( a \) and indent \( b \) and their topographical maps are presented in figure 7. The pile up around each indent indicates where plastic deformation has occurred. When indenting into a basal plane there is little out of plane displacement (a peak height of approx. 150 nm from the free surface) extending fairly equally around the indent impression. When indenting into a prism plane, the out of plane displacement is significantly higher (a peak height of approx. 400 nm from the free surface) extending in two distinct lobes above and below the indenter impression.
6. Simulation results

Simulations have been carried out on both the prismatic plane and the basal plane orientations, as in the experiments. Two groupings of slip systems have been assessed in the analyses, namely a-type: basal \( \langle a \rangle \), prismatic \( \langle a \rangle \) and pyramidal \( \langle a \rangle \); and \( \langle c + a \rangle \) type: basal \( \langle a \rangle \), prismatic \( \langle a \rangle \), pyramidal \( \langle a \rangle \) and pyramidal \( \langle c + a \rangle \). The results obtained when only considering \( \langle a \rangle \) type slip are compared with those obtained when considering both \( \langle a \rangle \) and \( \langle c + a \rangle \) type slips in the simulations.

(a) Load–displacement curves

The computed load–displacement curves are compared with the experiments in order to determine the CRSS used in the crystal plasticity model. A range of CRSSs were tested. When the CRSSs were chosen as 210 MPa for \( \langle a \rangle \) type slip
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and 840 MPa for \( \langle c + a \rangle \) type slip (which is about the same value as reported by Viswanathan et al. (2005)), the simulated load–displacement curves show very good agreement with the experimental data when considering both \( \langle a \rangle \) and \( \langle c + a \rangle \) type slips in the analysis, as shown in figure 8. For comparison, both the load–displacement curves for indent \( a \) and indent \( b \) are shown in figure 8, and it can be seen that the load developed for indent \( a \) (on the basal plane) is larger than that for indent \( b \) (on the prismatic plane) to produce the same indentation displacement.

Figure 8 also shows that the predicted load is larger when considering only \( \langle a \rangle \) type slip than when both \( \langle a \rangle \) and \( \langle c + a \rangle \) type slips are included. This is found to apply for both indents (into prismatic and basal planes). This results from the different Schmid factors for \( \langle a \rangle \) type and \( \langle c + a \rangle \) type slip in these particular crystal configurations. Although the CRSS is smaller, the Schmid

factor is also much smaller for $\langle a \rangle$ type slip than that for $\langle c + a \rangle$ type slip, which strongly inhibits $\langle a \rangle$ type slip. For indent $a$, the $\langle a \rangle$ type slip directions are near-perpendicular to the loading direction, so that the Schmid factor for all $\langle a \rangle$ type slip systems is near-zero. As a result, a larger load is needed to generate sufficient resolved shear stress to activate $\langle a \rangle$ type slip systems. For indent $b$, the basal $\langle a \rangle$ slip plane is near-parallel to the loading direction, so that the Schmid factor for the basal $\langle a \rangle$ slip systems is very small and it is difficult for these slip systems to become activated; the only $\langle a \rangle$ type slip systems which can be activated are
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Figure 7. Topographical AFM maps of indents of similar orientation to indents a and b: (left) into the basal plane; (right) into the prism plane. Scale bar, 5 μm.

Figure 8. Load–displacement curves obtained from experiments and simulations of indent a, basal, and indent b, prismatic. Dot dashed line, experiment A; filled circle, simulation A \{c + a\} and \{a\}; open circle, simulation A \{a\} only; solid line, experiment B; filled square box, simulation B \{c + a\} and \{a\}; open square box, simulation B \{a\} only.

prismatic \{a\} and pyramid \{a\} slip systems. Given the crystal orientation in indent b, the Schmid factors for these slip systems are much smaller than those for \{c + a\} type slip and therefore the load needed to generate sufficient resolved shear stress to activate them is larger than that for the \{c + a\} type slip systems.

(b) Material displacement pile-up patterns

Figure 9 shows simulated material pile-up patterns (out of plane displacements) for indent a, basal, and indent b, prismatic, when considering only \{a\} type slip and when considering both \{a\} and \{c + a\} type slips. The pattern obtained when
considering just \( \{a\} \) type slip is quite different to that when considering both \( \{a\} \) and \( \{c + a\} \) type slips for indent \( a \), indicating that \( \{c + a\} \) slip plays significant part in allowing material pile-up when indenting into the basal plane. For indent \( b \), the patterns obtained from the two simulations are near-identical and it can be argued that \( \{a\} \) type slip dominates in material pile-up when indenting into the prismatic plane. The peak of the pile-up is just over approximately 70 nm high when indenting into the basal plane and approximately 300 nm when indenting into the prismatic plane, when considering \( \{a\} \) and \( \{c + a\} \) type slips.

(c) Lattice rotations

Predicted lattice rotation patterns on both the top indented surface and the midsection of the indented crystal are presented and discussed for both indent \( a \) and indent \( b \). Figures 10 and 11 show the predicted lattice rotations on the top surface for indent \( a \), basal, and indent \( b \), prismatic, respectively. In figure 10, for basal plane indentation, the two plots in the first row are the rotations about the \( X_3 \)-axis. Three pairs of positive and negative rotation lobes can be seen when considering \( \{a\} \) and \( \{c + a\} \) type slips, giving an approximate threefold rotational symmetry about the \( X_3 \)-axis, in reasonable agreement with
Figure 10. Predicted lattice rotation fields on the $X_1X_2$ plane around indent $a$. Columns: left, considering $\langle a \rangle$ and $\langle c + a \rangle$ type slip; right, considering $\langle a \rangle$ type slip only; rows: top, $\omega_{12}$; middle, $\omega_{23}$; bottom, $\omega_{31}$. (Colour scale is in radians.)

the observed experimental measurements shown in figure 5. The rotation pattern is slightly different when considering only $\langle a \rangle$ type slip, although there are also three pairs of positive and negative lobes but these do not show the same rotational symmetry about the $X_3$-axis. The agreement with experiments is significantly poorer in the absence of the inclusion of $\langle c + a \rangle$ type slip. The two plots in the second row are the rotations about the $X_1$-axis. The rotations on the
two sides of the indentation (either side of the $X_1$-axis) can be seen to be opposite in sign in both simulations, resulting from the symmetry of the indenter geometry, which is consistent with the experimental observation. The last two plots in figure 10 show the rotations about the $X_2$-axis. Similarly, the predicted rotations either side of the $X_2$-axis are of opposite sign in both simulations, resulting also from the indenter geometry, and consistent with the experimental observations.

Figure 11 shows the simulated lattice rotations on the top surface of indent $b$, into the prism plane, in the same order as in figure 10. There is no obvious rotational symmetry predicted about the $X_3$-axis when considering both $\langle a \rangle$ and $\langle c + a \rangle$ type slips. For the rotation about the $X_1$-axis, both simulations give similar results, with rotations either side of the $X_1$-axis of opposite sign but this is not in obvious agreement with the experimental measurements shown in figure 4, which in themselves are not intuitive, given the indenter symmetry. For the rotation about the $X_2$-axis, both simulations give similar results but, again, show slightly different trends compared with the experiments.

Although it is apparent that there is some qualitative agreement between the predicted and experimental results, there are clearly some quite significant differences and the predicted results generally show a larger magnitude of lattice rotation than the experiments. In the nanoindentation simulation carried out by Zaafarani et al. (2006), similar sorts of differences were shown between crystal plasticity predictions and experiments. The predicted lattice rotations on the cross-sectional planes under a conical indenter were found to be larger in magnitude than the experimental results and also the simulated rotation patterns were found to be more symmetric compared to the experimental results. Here, however, only the top surface lattice rotations are available in the experiments and a potential factor leading to the differences between the simulations and the experiments is of course the indenter-sample contact behaviour; we have assumed zero friction between the two. An additional explanation for the over-predicted lattice rotations may rely on the simplification stated earlier; SSD density is assumed not to evolve with deformation and certainly for the development of immobile SSDs, the hardening which results is therefore under-predicted which may facilitate larger lattice rotations than would be observed in practice. The establishment of data for SSD evolution would enable this argument to be tested.

$(d)$ GND distributions

The predicted GND distributions on the top surface when considering both $\langle a \rangle$ and $\langle c + a \rangle$ type slips for both indents are shown in figure 12. A larger magnitude of GND density can be seen around indent $a$ than that around indent $b$. It can also be seen that the GNDs with high density are only confined to regions near the indent, which is consistent with the experimental observation of Viswanathan et al. (2005) and which indicates that only the region local to the indent carries high plastic strain gradient.

7. Discussion

There are correlations between the orientations of the titanium crystals, specifically the declination angle, and both the elastic and the plastic behaviour during nanoindentation. When the indentation load is applied parallel to the $c$-axis the material is both stiffer and harder.

Variations in stiffness are due to the different positions of Ti atoms within the hexagonal lattice; as the material is indented, load will be applied to different configurations of atoms and will resolve onto different combinations of atomic bonds. Zarkades & Larson (1970) demonstrated this variation as a function of orientation, by considering the anisotropy in the stiffness and compliance.
constants (as calculated by Fisher & Renken (1964)) for simple tensile tests, which resulted in a similar trend as the variation of stiffness with declination angle seen in figure 2.

The variation of modulus at a particular orientation (or within a single grain) can be accounted for by considering the complex stress state directly under the indenter and the size and shape of the elastic zone. As the subsurface microstructure has not been characterized, it is more than likely that some of the elastic zones have interacted with the second grain’s subsurface (by using a large grain size and discounting indents within 20 μm of a grain boundary on the sample surface, this should be minimized). Figures 4 and 5 show that the elastic strain fields extend a considerable distance (approx. 10 μm) from the centre of the indent even when unloaded, while the plastic deformation, revealed by lattice rotations, extends less far. The larger scatter shown in indentation modulus compared to hardness is in accordance with this. These two problems highlight difficulties in isolating stiffness measurements required to make direct calculations of the individual stiffness and compliance constants.

Hardness is a measure of the ability for a material to plastically deform under the indenter and thus directly depends on orientation of the Ti crystal due to the differences in critically resolved shear stress of the different slip systems involved. \( \langle c+a \rangle \) type slip is approximately three times as difficult to initiate as \( \langle a \rangle \) type slip (Viswanathan et al. 2005). However, as \( \langle a \rangle \) type slip can only deform material parallel to the basal plane, as the crystal is rotated, \( \langle c+a \rangle \) type slip will be required more for plastic deformation. In the extreme case where the load is applied directly parallel to the \( c \) direction (similarly to indent \( a \)), only motion of \( \langle c+a \rangle \) dislocations can accommodate the indentation by generating displacements into the surface of the crystal. This is demonstrated with the shape and extent of the patterning of \( \langle a \rangle \) type versus \( \langle c+a \rangle \) type dislocations around indent \( a \). While there are approximately 10 times as many \( \langle a \rangle \) type dislocations overall, they are located almost exclusively very close to the indenter edges, whereas the \( \langle c+a \rangle \) type dislocations extend a greater distance from the indenter, presumably from the upheaval of material subsurface resulting in the storage of \( \langle c+a \rangle \) threading dislocations into six distinct lobes.
AFM topographic maps, figure 7, show that the out of plane displacement of material around the indent (pile-up) is significantly different, both in extent and in shape, between indents made into a basal plane and a prismatic plane. This can easily be explained as \((c + a)\) type dislocations are typically harder to move and generate (their CRSS is higher) and therefore any indentation into a near basal plane would require many dislocations of this type to displace material out of the surface, which corresponds to both an increase in hardness and a small amount of pile-up. The qualitative differences in the shapes between these two indents can be accommodated by the underlying symmetry of the crystals, where by the material displaced, and is symmetric around the \(c\)-axis. One must be careful in interpreting both AFM images and GND maps together as they show wholly different phenomena: AFM images show where material has moved by the progress of dislocations; GND maps show where threading dislocations are being stored (which may be a result of deformation). These topographic maps compare well with the results from simulations as presented in figure 9. The anisotropy in these topographic maps illustrate some difficulties when attempting to interpret the size and shape of the plastic zone which surrounds the indenter; one must be careful not to over-interpret what the technique actually shows.

To confirm the effect of orientation and the nature of sub-surface deformation processes below the indent, FIB-SEM and EBSD tomography could be utilized to observe the lattice rotations and curvatures beneath the indent. Furthermore, by using a serial sectioning technique similar to Demir et al. (2009) combined with high-resolution EBSD analysis it should be possible to consider all the components of the deformation tensor and the gradients in three dimensions, which would provide a full calculation of the lattice curvature components and provide a less under-determined solution to equation (4.5). Using the analysis above, it would then be possible to separate the stored GNDs into \(a\) type and \((c + a)\) type dislocations, which could provide further information as to the nature of the deformation process and assist in the development of novel titanium alloys. Yet, there remain problems with three-dimensional FIB-EBSD tomography; each fresh surface will result in some elastic (and possibly plastic) relaxation, which will require comparison of EBSD results with simulations. Furthermore, the difficulties in very precise alignment and cutting of each slice will result in varying errors in the measured curvatures, making the whole reconstruction somewhat taxing (as demonstrated by the different apparent dislocation densities reported by Demir et al. (2009) with a five slice reconstruction versus a full volume reconstruction).

Overall spatial variations in the dislocation densities observed around indents \(a\) and \(b\) illustrate the importance of considering which slips systems will be active during indentation and thus how to accurately understand indentation results. Thus far this has been largely overlooked when applying simple Johnson cavity model approaches such as used in the Nix & Gao (1998) model.

One can evaluate the lowest GND density measurable by this technique by examining GND densities away from the indent, which constitutes well annealed material. The noise in the total GND density compared to the total \(a\) and \((c + a)\) type GND densities are slightly different, as noise is introduced upon measuring the lattice rotations (and thus curvatures) which then are processed.
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to calculate the GND densities on each individual slip system. Thus the noise floor is determined by a combination of the number of different slip systems presented in each map. For the total GND density this constitutes a lower bound of approximately $10^{13}$ GNDs m$^{-2}$ compared to approximately $10^{12}$ for the sum of $(c + a)$ type dislocations and approximately $10^{13}$ for the sum of $(a)$ type dislocations. We believe that our lower bound solution for GND solution is relatively stable, as when examining the GND densities of individual slip systems around the indent, one can notice clustering of high GND content for neighbouring points in the maps. As the GND density at each individual point in the map is solved individually and if the solution was unstable, the curvatures would likely be supported by a more varied combination of GND densities for neighbouring pixels within the map.

The out of plane displacements presented experimentally in figure 7 show considerable differences between material pile-up for indents $a$ and $b$, which can be qualified by the ease of $(a)$ type slip in indent $b$, resulting in the two lobes above and below the indenter impression and the relative difficulty of $(a)$ type slip for indent $a$ to accommodate pile-up. These displacements compare favourably well with the predicted displacements presented in figure 9.

Comparisons of predicted and experimentally determined lattice rotations show promise. Particularly, the lattice rotation features associated with indentation into the basal plane (indent $a$) were reasonably well captured. Interestingly, lattice rotations into the prismatic plane (indent $b$) were less well captured, though some aspects of the experimental observations were not intuitive. For both indent types, the magnitudes of the lattice rotations were over-predicted by the model. There are a number of reasons for this which equally should be taken into account when assessing the predicted rotation distributions as well as magnitude. Firstly, the model assumes that there is no multiplication of statistically stored dislocations with deformation so that the only material hardening allowed for comes from the generation of GNDs. The hardening predicted by the model is therefore likely to be lower than that in reality and this will lead to over-predicted lattice rotation magnitudes. A further clear difference between the simulated indentations and the experiments is that the withdrawal of the indenter, which was carried out in the experiments, was not accounted for in the simulations. Therefore the resulting stress relaxation in the experiments was not accounted for in the simulations and it seems intuitive that the resulting relaxation would lead to the reduction of lattice rotation, which is not captured in the modelling.

One further aspect of the modelling should be noted. The computer CPU times for the fully three-dimensional analysis were prohibitive. For this reason, the size of the single crystal modelled was considerably smaller than that in the experiments, and this raised a number of problems. Firstly, the boundary conditions chosen, as discussed above, were unlikely to represent exactly what was applied in the experiments. Secondly, as indentation proceeds, so the plastic zone size increases and it was found in the simulations that towards the end of the indentation, the plastic zones were starting to interact with the single-crystal free surfaces thereby generating boundary effects. This is seen in figure 11, for example, as the development of bands of lattice rotation extending from the indent to the free surfaces. A larger model would enable these effects to be reduced but at a cost of significantly higher CPU times.
Finally, as stated earlier, the model simulations assume frictionless conditions between the indenter and the Ti single crystal. While other simulations of indentation have shown that the effect of friction is limited, this remains to be validated by experiment.

8. Conclusions

The combination of high-resolution EBSD and crystal plasticity modelling shows much promise in understanding fundamental deformation processed both during indentation of small volumes and within complex engineering materials.

High-resolution EBSD is an emerging SEM-based technique which spectacularly furthers the potential of a now common laboratory tool. Material scientists and engineers now have the ability to conduct precise measurement of strain and rotation within realistic volumes of material, thus bridging length scales between typical large-scale microscopy techniques such as X-ray and small-scale techniques such as TEM. Here we have used the technique to evaluate a lower bound solution for the density of stored GNDs, resolved onto individual slip systems, to examine deformation processes in a manner more applicable to real engineering applications.

We have used nanoindentation to probe small single crystals of titanium of varying crystal orientation and demonstrated the fundamental variation of mechanical properties. We have presented some visual maps of the varying volumes of material which are actually probed during indentation and must be considered when utilizing this technique on real materials.

Crystal plasticity model simulations were shown to capture correctly the indentation load–displacement characteristics for both basal and prismatic indentation. The lattice rotation fields resulting from basal indentation were well captured, particularly the threefold symmetry in the rotations about the axis parallel with the indentation direction. It was found that it was essential to incorporate \( \langle c + a \rangle \) type slip in the model in order to capture this effect, because of the need for pyramidal slip to accommodate the plastic deformation with this configuration. As with other indentation models, the predicted magnitudes of lattice rotation were higher than those observed experimentally.

We have combined nanoindentation, high-resolution EBSD and crystal plasticity model simulations to accurately examine the mechanical properties on titanium and validate many of our assumptions.

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