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Deformation behaviour of [001] oriented MgO using combined *in-situ* nano-indentation and micro-Laue diffraction

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ABSTRACT

We report a coupled *in-situ* micro-Laue diffraction and nano-indentation experiment, with spatial and time resolution, to investigate the deformation mechanisms in [001]-oriented single crystal MgO. Crystal plasticity finite element modelling was applied to aid interpretation of the experimental observations of plasticity. The Laue spots showed both rotation and streaking upon indentation that is typically indicative of both elastic lattice rotation and plastic strain gradients respectively in the material. Multiple facets of streaking of the Laue peaks suggested plastic slip occurring on almost all the {101}-type slip planes oriented 45° to the sample surface with no indication of slip on the 90° {110} planes. Crystal plasticity modelling also supported these experimental observations. Owing to asymmetric slip beneath the indenter, as predicted by modelling results and observed through Laue analysis, sub-grains were found to nucleate with distinct misorientation. With cyclic loading, the mechanical hysteresis behaviour in MgO is revealed through the changing profiles of the Laue reflections, driven by reversal of plastic strain by the stored elastic energy. Crystal plasticity simulations have also shown explicitly that in subsequent loading cycles after first, the secondary slip system unloads completely elastically while some plastic strain of the primary slip reverses. Tracking the Laue peak movement, a higher degree of lattice rotation was seen to occur in the material under the indent, which gradually decreased moving laterally away. With the progress of deformation, the full field elastic strain and rotation gradients were also constructed which showed opposite lattice rotations on either sides of the indent.

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1. Introduction

MgO is an interesting ceramic material owing to the fact that it is deformable at room temperature by conventional slip processes. Since the demonstration of this feature by Hulse et al., MgO has been a material of interest for several decades [1]. In spite of having a large intrinsic strength characteristic of a ceramic material, with a NaCl-type structure, MgO presents a model system to understand plasticity in ceramics that have more than five independent {110}110 -slip systems. Hulse et al. showed that single crystal MgO exhibited appreciable compression ductility at all temperatures from −196 °C to 1200 °C [1,2]. Single crystal deformation of MgO

has been tested earlier though indentation, tension, bulk and micropillar compression [1–6]. Post-deformation determination of deformation mechanisms have been carried out primarily by electron microscopy [7,8], chemical etching [9] and atomic force microscopy [10]. Microindentation studies on (001)-MgO surface showed the formation of shear-induced microcracks that developed due to misorientations in the periphery of the indent – no dislocation structures consistent with the dislocation pile-up model for crack was observed [8]. As a result, nano-indentation experiments were performed to study plasticity at low stress regimes [10] and again deformation mechanisms were described based on observations of dislocation movement by etch-pits and scanning probe microscopy [11]. In addition MgO exhibits a high mechanical anisotropy resulting from the difference in critical resolved shear stresses in primary and secondary slip systems – as a result of this anisotropy, it shows reversible plastic flow upon

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cyclic loading [12]. This mechanical anisotropy typically manifests as hysteresis loop in load-displacement plot. While recent literature has reported observation of this hysteresis, no clear explanation of the structural variation during the reversible flow has so far been established.

The combination of nano-indentation and time-resolved *in-situ* micro-Laue diffraction could be effective in correlating the measured mechanical properties with the microstructural evolution, e.g. critical stresses for activation of slip systems, density of geometrically necessary dislocations (GND), and creation of subgrains. Recently, *in-situ* micro-Laue diffraction has been used to study detailed material response under various forms of loading such as tensile [13], micropillar compression [14–18] and bending [19]. The technique has been used on a range of materials including pure metals, two-phase metallic alloys, complex metallic alloys and ceramics. In Laue diffraction, the position of the Laue spots is dependent on the orientation of the crystal and lattice strain [20]. Changes in the crystal orientation, lattice strain and lattice strain gradients manifests as changes in the shape and position of the peaks. For example, Barabash et al. suggested that from direct measurement of the aspect ratio of streaking, caused due to slip on a given slip system, the dislocation density can be estimated [21]. Also with applied deformation, a given Laue reflection can split into two or more peaks and the angular relationship between the split peaks essentially represents a subgrain boundary within a parent volume. This means that analysis of peak splitting can be used to form an idea of the population of dislocations present within a misorientated subgrain and those that form the subgrain boundary [22]. Thus far, micro-Laue experiments have been coupled with micropillar compression, both *in-situ* and *ex-situ*, to understand the distribution of stress [23], effect of prior defects in materials [24,25] and activation of slip processes [17,18]. By using coupled *in-situ* micropillar and micro-Laue experiments, a detailed time-resolved analysis of the activation of slip sequentially on multiple systems in pure metals have been reported [17,18,26]. Through such experiments supported by crystal plasticity modelling, even the early initiation of slip on other slip systems, *a priori* expected slip systems, activated due to pre-existing strain within the sample volume or pillar shape, has been convincingly captured [14,24,27]. Also, by scanning the volume of a deformed micropillar by a focused sub-micron beam, a spatial map of the rotation between various sections of the pillar can be evaluated, and furthermore using the extent of rotation a full description of the geometrically necessary dislocations can be derived [23]. In recent *in-situ* bending experiments on gold nanowhiskers, the elastic properties could be extracted through rotation of the Laue spots [19].

The stress state under a Berkovich indenter is complex and this has a pronounced effect on the deformation behaviour of the material underneath. Analysing the evolution of the stress-field can help explain the elastic and plastic flow of the material under the indent. Using a sub-micron sized probe can help spatially resolve and quantify the deformation gradient in the material under a nano-indenter. Recently, using a monochromatic probe, the complex elastic strains generated beneath a nano-indenter, along the transverse and depth directions, in a Zr-bulk metallic glass have been reported [28]. Nano-indentation can also potentially offer a unique scope to perform mechanistic study of the phenomenon of reversible hysteresis in single crystal MgO, as discussed earlier, through the development of a complex state of strain that in turn would incite slip on both primary and secondary slip systems. Using a uniaxial loading this would have necessitated a polycrystalline sample with “hard” and “soft” grains – in that case there would have been an added complexity introduced from the presence of grain boundaries in the samples. We believe that by combining nano-indentation with time-resolved Laue diffraction

experiment, detailed analyses of the micromechanisms behind the mechanical hysteresis in MgO can be made.

Through this work, using a white beam *in-situ* micro-Laue diffraction we aim to study:

- i) the slip processes that occur beneath a nano-indenter in a MgO single crystal;
- ii) the elastic deformation gradient that exists within the material both as a function of time, applied load and distance from the indenter; and
- iii) the mechanisms of reversible hysteresis during cyclic loading in MgO.

We hope that these results can be used to understand other mechanically anisotropic materials exhibiting a reversible hysteresis behaviour as a result of residual stored elastic energy.

2. Experimental procedure

Commercially available [001]-oriented MgO single crystal was sectioned and mechanically thinned down to 150 μm thickness. Cylindrical pillars of dimensions 10 μm (diameter) $\times$ 10 μm (height) were milled on the thin edge of the sample, with the pillar parallel to [001], using focused ion beam (FIB) on a Helios NanoLab under an accelerating voltage of 30 kV and final current of 90 pA. Tapering of $\sim 1^\circ$ due to FIB machining was likely present in the pillars, but this is unlikely to affect our interpretation of the deformation fields. Since the experiments were to be performed in transmission geometry, significant areas around the pillars were also milled out in order for the transmitted beam to pass through uninterrupted. The *in-situ* micro-Laue experiments were performed at beamline 34-ID-E in Advanced Photon Source, Argonne National Laboratory, USA on a custom-built Alemnis loading rig. The pillars were indented under a load-controlled mode cyclically twice using a Berkovich tip with a loading (and unloading) rate of 0.2 mNs^{-1} up to a peak load of 25 mN. As the sample was indented white light with energy ranging from 7 to 30 keV was shone with the incident beam focused to 0.3 $\mu\text{m} \times 0.3 \mu\text{m}$ with a pair of Kirkpatrick-Baez (KB) mirrors. A schematic of the experimental set-up is shown in Fig. 1. In order to build up a three-dimensional picture of the strain under the indenter, the incident spot was made to raster the micropillar in a 2×5 array with a step size of 2 μm near the top of the pillar. The points of data acquisition, as numbered, are shown in Fig. 1. In this way, both time and spatially resolved map of deformation under an indent could be obtained. The Laue patterns were captured during the experiment using a 165 mm diameter Mar165 CCD camera, containing a 61 mm $\times$ 61 mm sensor with 80 μm pixels, at an exposure time of 3 s – including the read-out time of around 7 s, the time to generate a frame was ~ 10 s. The sample-detector distance was 114 mm. This was worked out by placing a standard Si-wafer in the beamline and calibrating with respect to indexed Laue reflections. Following indentation, the pillar was cross-sectioned and using the *in-situ* lift-out technique in a FEI Helios NanoLab scanning electron microscope (SEM), the deformation microstructure was studied with a JEOL 2100F transmission electron microscope (TEM).

3. Crystal plasticity finite element modelling

Crystal plasticity finite element (CPFE) models were used to study the deformation behaviour during the indentation in this study. The crystal plasticity model can predict the slip behaviour in the individual slip systems, which will be used to explain the observations made during the experiment.

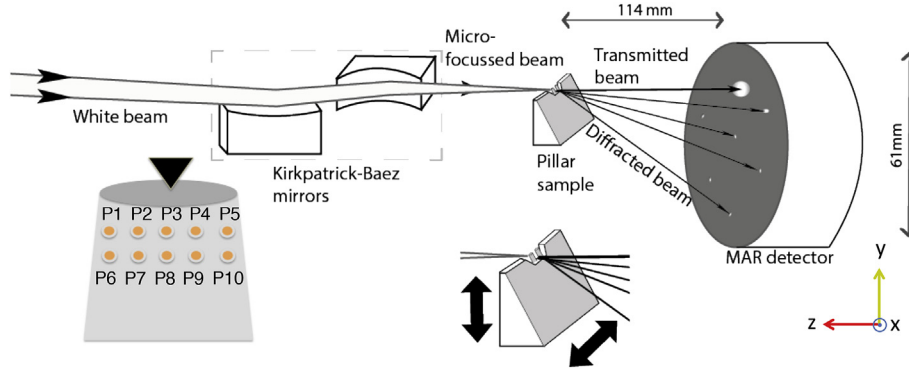


Fig. 1. Schematic of the micro-laue set-up used during the experiment at beamline 34-ID-E, Advanced Photon Source. The incident white beam had an energy range of 7–30 keV. The sample containing the pillars was mounted on the Alemnis compression rig. The thick double-headed arrows show the direction of movement of the sample stage to raster the beam across the whole volume of the pillar as shown.

3.1. Crystal plasticity material model

3.1.1. Constitutive equation

The crystal plasticity model used in this study is based on the work done by Asaro [29], and the constitutive equations of the crystal plasticity are summarised as follows,

$$\tau^\alpha = m_i^\alpha \sigma_{ij} s_j^\alpha \quad (1)$$

$$\dot{\gamma}^\alpha = \dot{\alpha} \cdot \text{sgn}(\tau^\alpha) \left(\left| \frac{\tau^\alpha}{g^\alpha} \right| \right)^n \quad (2)$$

$$\dot{g}^\alpha = \sum_\beta h_{\alpha\beta} \dot{\gamma}^\beta \quad (3)$$

$$\dot{\sigma}_{ij} = C_{ijkl} (\dot{\epsilon}_{kl} - \dot{\epsilon}_{kl}^p) = C_{ijkl} \left(\dot{\epsilon}_{kl} - \sum_\alpha \frac{1}{2} \dot{\gamma}^\alpha (s_k^\alpha m_l^\alpha + s_l^\alpha m_k^\alpha) \right), \quad (4)$$

where τ is the resolved shear stress, m_i and s_j are the slip plane normal and slip direction respectively, g the slip system hardness, γ is the plastic shear strain, $\dot{\alpha}$ and n are material parameters, h is the slip plane hardening moduli, σ is the stress, ϵ is the strain, and C the fourth-order stiffness tensor. The subscripts and superscripts α and β denote the α -th and β -th slip systems respectively. The number of slip systems and their orientations depend on the crystal lattice structure. Lastly, the superscript p denotes plastic strain and the dot above the variable denotes the time derivative (i.e. the rate) of the variable.

The hardening laws that describes the values of $h_{\alpha\beta}$ were based on the work by Asaro [29] and Peirce et al. [30], which are as follows.

$$h_{\alpha\beta} = \begin{cases} h(\gamma) = h_0 \text{sech}^2 \left| \frac{h_0 \gamma}{\tau_s - \tau_0} \right|, & \alpha = \beta \\ qh(\gamma), & \alpha \neq \beta \end{cases} \quad (5)$$

$$\gamma = \sum_{\alpha=1}^n |\gamma^\alpha|, \quad (6)$$

where $h_{\alpha\alpha}$ (or $h_{\beta\beta}$) and $h_{\alpha\beta}$ are the self and latent hardening moduli respectively, h_0 is the initial hardening gradient, τ_0 is the initial critical resolved shear stress, τ_s is the saturated critical resolved shear stress, and q is the hardening factor, which is set to unity

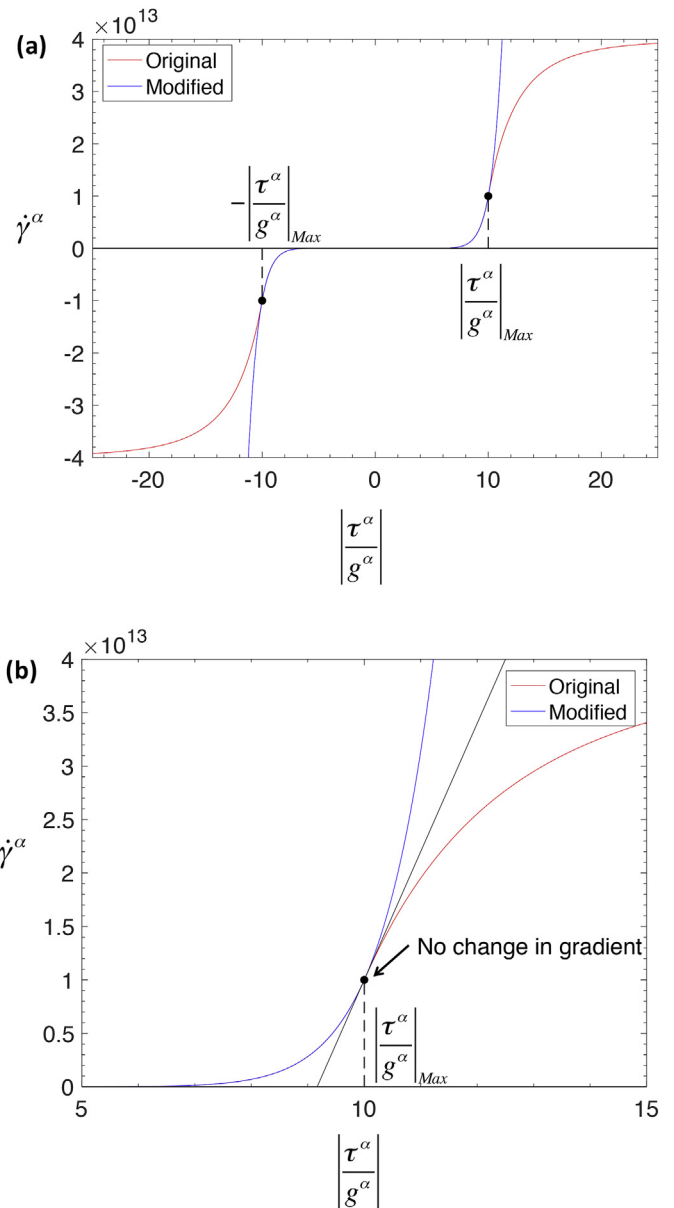


Fig. 2. Shear strain rates vs resolved shear stress curves comparing the strain rates using Eq. (2) and the original equation. (a) Full curve (b) Zoomed in portion showing gradient at $|\tau^\alpha/g^\alpha|_{Max}$.

assuming Taylor's isotropic-hardening.

The crystal plasticity material model is implemented in a commercial finite element software, Abaqus, via a modified UMAT subroutine from Huang [31]. Since the shear strain rate in Eq. (2) increases exponentially with $\left|\frac{\tau^\alpha}{g^\alpha}\right|$, as seen in Fig. 2(a), which causes convergence problems with the subroutine Eq. (2) is modified as follows.

$$\dot{\gamma}^\alpha = \begin{cases} \dot{\alpha} \cdot \text{sgn}(\tau^\alpha) \left(\frac{\tau^\alpha}{g^\alpha} \right)^n, & \left| \frac{\tau^\alpha}{g^\alpha} \right| \leq \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}} \\ \left[B_1 + B_3 \left| \frac{\tau^\alpha}{g^\alpha} \right| + \left(\frac{B_2}{1-B_3} \right) \left| \frac{\tau^\alpha}{g^\alpha} \right|^{1-B_3} \right] \cdot \text{sgn}(\tau^\alpha), & \text{otherwise} \end{cases} \quad (7)$$

where B_3 and B_4 are set to give a smooth cap to the strain rate when $\left| \frac{\tau^\alpha}{g^\alpha} \right| > \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}}$, while B_1 and B_2 are calculated as follows,

$$B_2 = \frac{\left(\dot{\alpha} n \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}}^{n-1} - B_4 \right)}{\left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}}^{1-B_3}} \quad (8)$$

$$B_1 = \dot{\alpha} \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}}^n - \left(\frac{B_2}{1-B_3} \right) \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}}^{(1-B_3)} - B_4 \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}} \quad (9)$$

The modified Eqs. (7)–(9) will produce a cap for the strain rate while keeping the gradient at $\left| \frac{\tau^\alpha}{g^\alpha} \right| = \left| \frac{\tau^\alpha}{g^\alpha} \right|_{\text{Max}}$ to prevent any discontinuities in the rate and its gradient.

Table 1
Material parameters used in CPFE model in this study.

Parameter	{101} < 10 $\bar{1}$ >	{001} < 110 >
τ_0 (MPa)	47.1	2000
τ_s (MPa)	65.66	2200
h_0 (MPa)	200	30
$\dot{\alpha}$	0.01	0.01
n	12	12
$\left \frac{\tau^\alpha}{g^\alpha} \right _{\text{Max}}$	5	5
B_3	−2.5	−2.5
B_4	0	0

3.1.2. Material properties

The material properties used in the CPFE analysis are that of a single MgO crystal. MgO is an orthotropic material and the values of the stiffness tensor for room temperature were obtained from Ref. [32], which are $C_{11} = 296.6$ GPa, $C_{12} = 95.9$ GPa, $C_{44} = 156.2$ GPa. In this work, two families of slip systems, {101} < 10 $\bar{1}$ > and {001} < 110 >, are denoted as the first and second families of slip systems respectively. The various material parameters used in the simulation are summarised in Table 1.

3.2. Finite element modelling and verification of experimental parameters

A finite element model was constructed to simulate the experiment. The MgO micropillar is modelled as a cylinder with 10 μm diameter and a height of 3 μm as shown in Fig. 3. The finite element model of the MgO was made entirely from linear hexahedral (C3D8R) elements, while the Berkovich indenter is treated as a rigid surface. The finite element model of the MgO was partitioned into two regions, a fine mesh region denoted as the core, as shown in Fig. 3, with a diameter of 4.5 μm and an external region with a coarser mesh. The core region has a total of 32896 elements and 34451 nodes with the material modelled using crystal plasticity UMAT subroutine. The external region, with 1568 elements and 9773 nodes, which is expected to not have significant plastic deformation, was modelled as an elastic material. Since the MgO is a single crystal, all elements were set to have the same material orientation, which is defined based on the global coordinate system shown in Fig. 3.

The bottom face of the MgO was set to have zero displacements while the Berkovich indenter was set to indent to a maximum depth of 355 nm at a constant rate of 0.2 nm s^{-1} , which yielded a load close to the peak load measured in the experiment and this was followed by the indenter returning to its original position at the same rate. The depth and rate of the indenter was based on the displacement readings obtained from the experiment. A total of two loading cycles were applied to the MgO in the simulation and the displacements and load of the indenter in the simulations are shown in Fig. 4(a). The peak load from the simulation (Fig. 4) was found to be 22 mN, which is close to the experimental value of 25 mN, suggesting that the finite element model is reasonable. This minor discrepancy is likely to be caused by the minor errors in the critical resolved shear stresses used in the simulations and the depth being slightly different in the experiments compared to the simulations. For the simulated load and displacement versus time curves, the corresponding load-displacement curves were obtained and presented in Fig. 4(b and c). The simulated curves were obtained for two loading cycles and showed the clear presence of

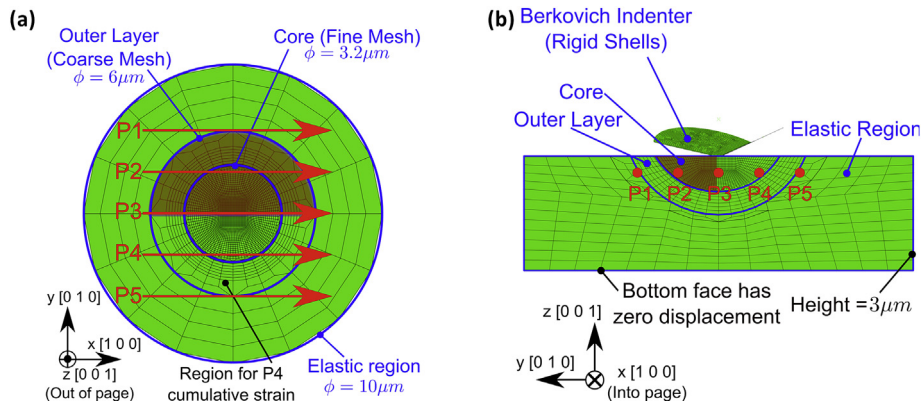


Fig. 3. A schematic of the finite element mesh showing the (a) top view and (b) side view of the cross section.

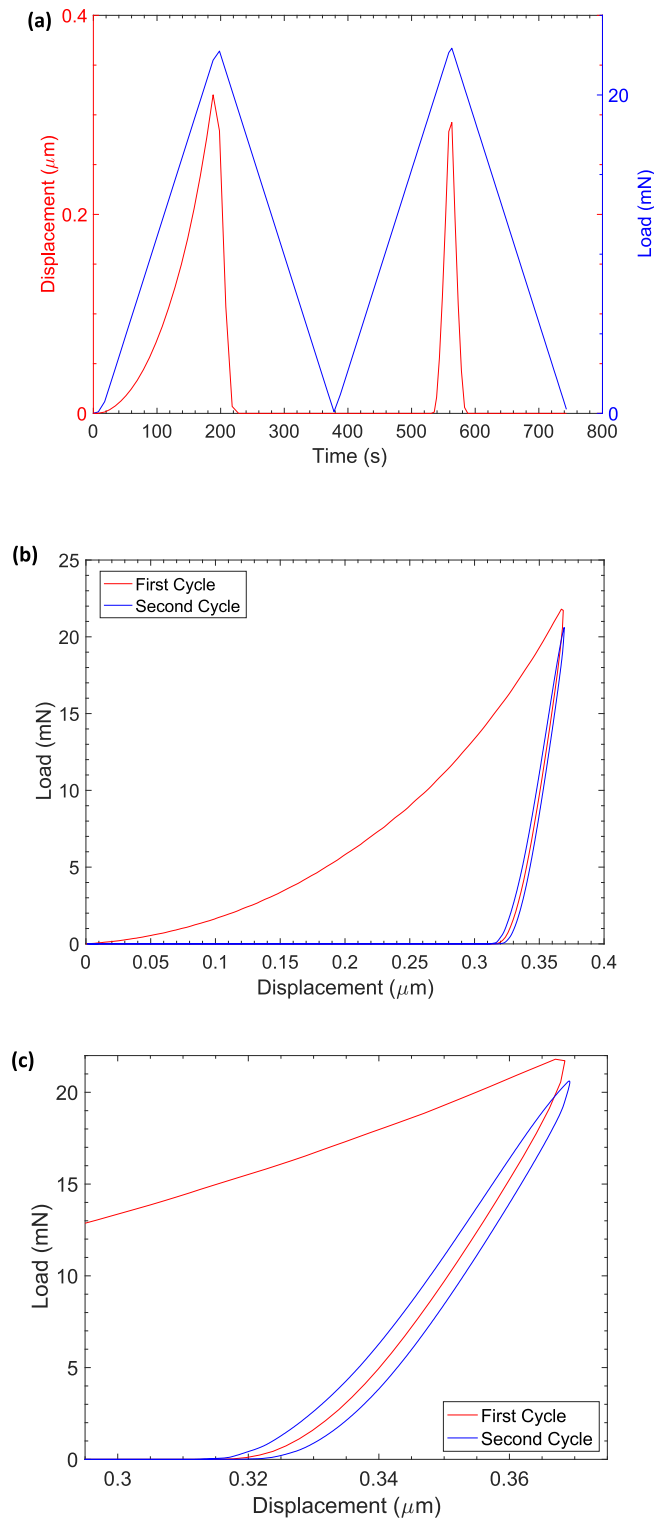


Fig. 4. (a) Simulated load and displacement of indenter with time and (b) simulated load-displacement curve showing the hysteresis in the second and third cycle. (c) Magnified section of the loading-unloading part.

reversible hysteresis. This is in line with the previously reported experimental work on cyclic indentation of MgO single crystals that showed reversible hysteresis [12].

4. Results and discussion

4.1. Influence of indentation load on Laue patterns

4.1.1. Time resolved patterns

An exemplary Laue pattern obtained from the MgO micropillar before indentation is shown in [Supplementary Fig. 1](#). From the pattern, the exact beam direction was obtained to be $[17\ 0\ 3]$, which is about $\sim 9.6^\circ$ away from the orthogonal $[100]$ direction. In order to study the evolution of deformation with time and indentation load, one peak from the whole Laue pattern was selected, from a specific location on the pillar, in this case P3, to track. For this, the $(1\bar{1}3)$ peak was selected for analyses. [Fig. 5](#) shows the changing appearance of the peak with respect to both time and applied load. Since the beam was scanned across the width of the pillar, the first frame was captured from position P3 when the pillar was already under a load. The width of the initial spot at the base was $\sim 0.9^\circ$. As mentioned, the streaked nature of the peak in part can be due to this applied loading or also an artifact arising from the FIB milling process. FIB milling is known to induce prior strain gradients within micropillars due to sputtering and radiation damage [24,33,34], which is dependent on both the size and material of the pillar. This existing strain gradient influences the plastic deformation of pillars at initial stages and results in an extraneous hardening before slip on geometrically predicted slip system can start operating. Pillars were milled with a 30 kV ion beam that typically induces 30 nm of damaged layer on the surface. A similar streakiness is also seen in other peaks which are examined from the same initial pattern from P3, as shown in [Fig. 6](#), the peaks plotted are $(1\bar{1}3)$, (115) , $(1\bar{3}3)$, (135) , $(1\bar{3}1)$ and $(1\bar{1}5)$. Focusing attention back to [Fig. 5\(a–h\)](#), we observed that with progressive indentation of the pillar, distinct changes in the shapes and positions of the peaks were noticed though the two loading cycles until complete unloading of the pillar ([Fig. 5\(h\)](#)). At first, during the first loading cycle, an immediate rotation of the peak is observed as the peak centre, marked by the cross hair, moves markedly ([Fig. 5\(a and b\)](#)). By taking into account the position of the $(1\bar{1}3)$ peak on the CCD detector and for the known sample-detector, a maximum rotation of 0.15° and 0.2° were calculated for the peak load after first and second cycles respectively. This movement of the peak is believed to occur as a result of elastic deformation (rotation and shear strain) induced in the material around the indent. Subsequently, there is also a distinct streaking of the peak when the load rises to almost the peak load. No abrupt changes in the path of rotation of the peaks, that might suggest discrete strain bursts within the sample caused by abrupt slip events like accumulation of dislocations or activation of a competing slip system, were apparent during indentation. With the removal of load the extent of streaking reduces gradually, and following unloading after the cycle the peak is still streaked and did not revert back to the initial position. The direction of this streaking bears an imprint of slip occurring on a particular slip system – this will be discussed in section 4.1.2. Under the influence of this particular slip system different peaks would be naturally expected to streak along different directions within the whole Laue pattern. This is also evident in [Fig. 7](#), where the different peaks have a distinctive sense of streaking. Upon loading for the second cycle, the peak again showed a gradual change of shape, as observed during the first cycle. The extent of streaking in the peak ([Fig. 5\(f\)](#)) up to the maximum load is also found to be similar. Following the end of the second cycle a residual streaking is present in the peak, which implies that the sample had undergone a plastic deformation. Also the peak does not completely revert back to the initial

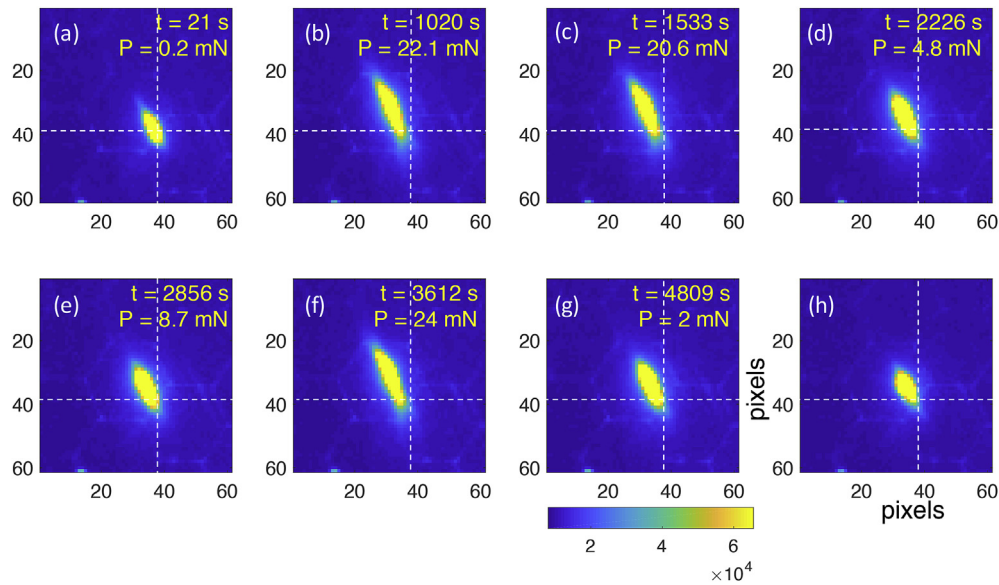


Fig. 5. (a–h) Evolution of the $(1\bar{1}3)$ reflection with increasing indentation load showing streaking and also minor rotation of the peak. The time and the indentation load are stamped on the frames. The cross-hair shows the position of the initial peak-centre. Pixel size is $80\ \mu\text{m}$. The intensities are in arbitrary units.

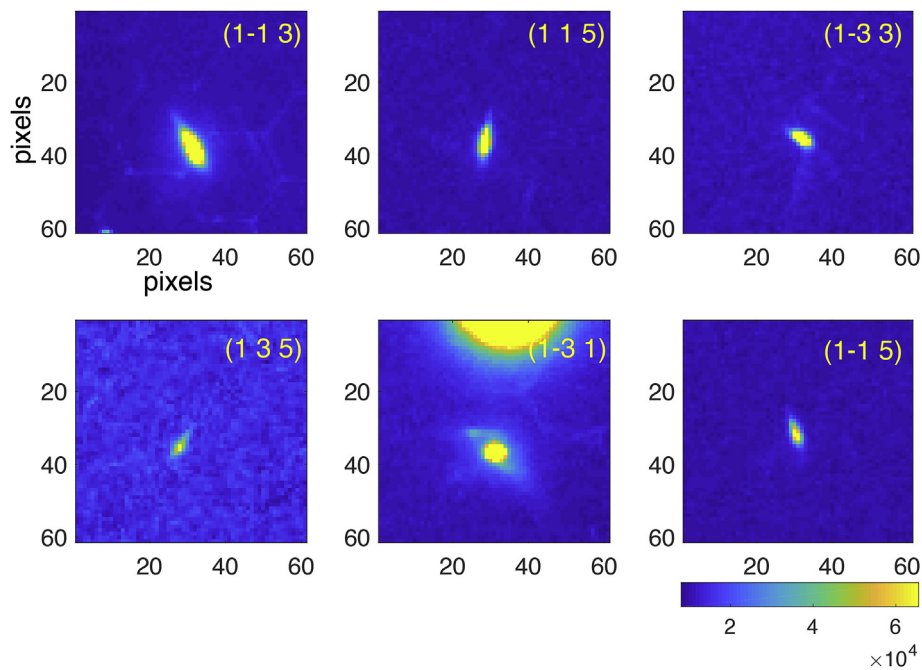


Fig. 6. Snapshots of different peaks obtained from the initial frame ($t=0\ \text{s}$) from position P3 showing an initial streaking observed in all the peaks. Pixel size is $80\ \mu\text{m}$. The intensities are in arbitrary units.

position implying a creation of distinct misorientation in the sample volume and again this misorientation corresponds to a rotation of 0.1° of the peak with respect to its initial position.

4.1.2. Spatially resolved patterns

As the white-beam was rastered across the indent, the deformation mechanism at different locations within the material, near to far from the indent, could be captured successfully, as shown in Fig. 8. Under a given load, the snapshots of the $(1\bar{1}3)$ peak from different locations of the pillar, where deformation under the indent was expected, were recorded and shown in Fig. 8. As can be

seen, moving inwards from the sides of the micropillar to the centre, the extent of streaking evidently increases. This is expected from the nature of strain gradients moving horizontally from the far field to the region under the indenter. Also, a similar reduction in intensity is seen along the vertical direction moving from the top towards the bottom of the micropillar, which shows a gradual reduction of the strain along the depth under the indent. A closer look at the streaking of the Laue spot from positions P3 and P4 particularly in Fig. 9 reveals that the senses of streaking of the peaks are in multiple directions. It is to be noted here that the position of the indenter lies between points P3 and P4 and this is slightly offset

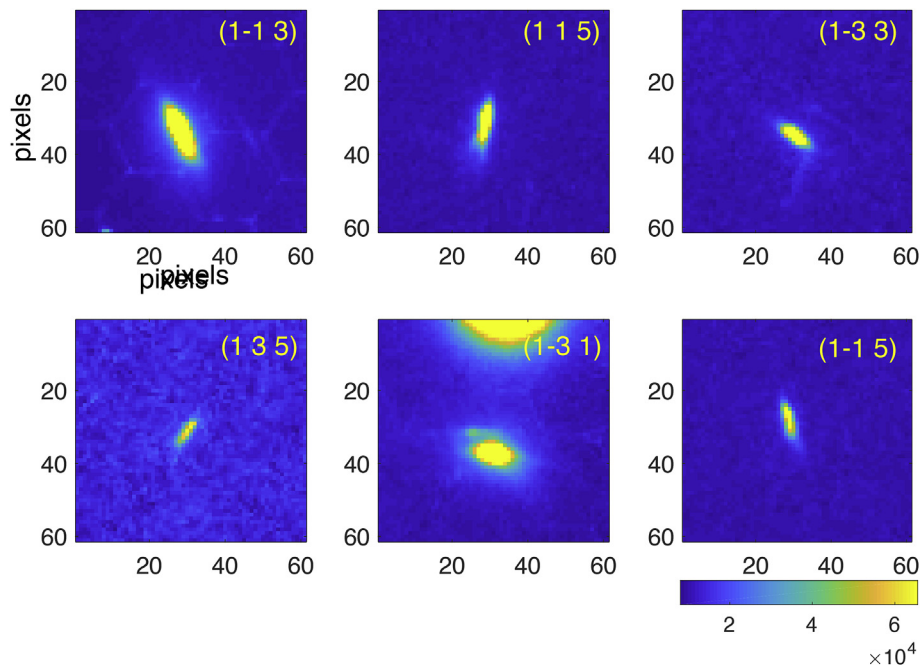


Fig. 7. Images of different Laue peaks obtained from the frame corresponding to the peak load of the first indentation cycle from position P3. The peaks show streaking along different directions under the effect of slip in the left half of the pillar. Pixel size is $80\ \mu\text{m}$. The intensities are in arbitrary units.

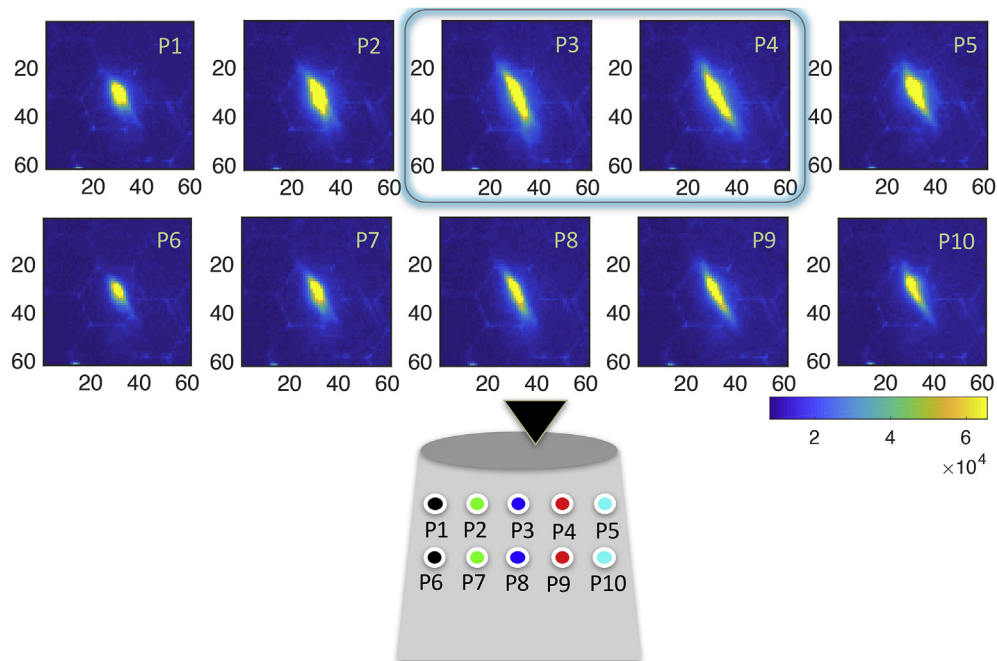


Fig. 8. Snapshot of the $(1\bar{1}3)$ peak obtained from the different locations of the pillar, as shown on the right with an exposure time of 3s. Also superimposed is the location of the indent with respect to the pillar axis based on the observation of the streaking of the peaks. The intensities are in arbitrary units.

from the exact centre of the pillar – this is quite reasonable since it is difficult to indent exactly the centre of the pillar during *in-situ* experiments such as these with limited visibility of the indent and the pillar. Persistent slip on a given slip system aided by the generation of an excess population of geometrically necessary dislocations gives rise to a deviatoric strain gradient in the sample volume and this manifests as continuous streaking of Laue peaks. The direction of streaking of a Laue spot, points in the direction of

the plane towards which the corresponding plane would rotate under the assumption of slip on a given slip system – in this way the streaking direction gives an indication of the slip system being activated in the material. Assuming a given slip system, a rotation axis $\mathbf{N}$, and angle, θ , both can be obtained from the cross-product and dot-product respectively of the known loading direction and slip plane normal. Using the rotation axis and the angle between the slip plane normal and loading direction (θ), a 3×3 Rodriguez

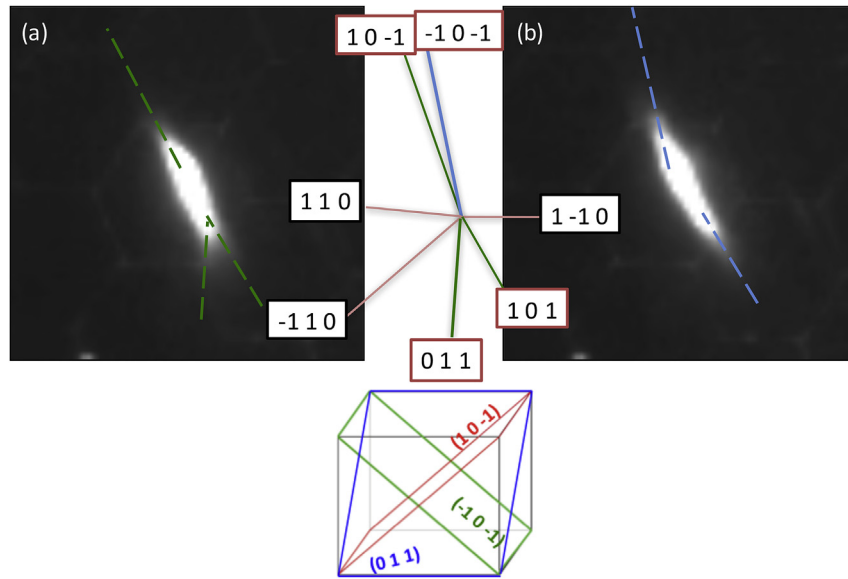


Fig. 9. The streaked reflections from spots (a) P3 and (b) P4 on which is superimposed the vectors denoting the direction which the spot would streak given slip on various $\{110\}$ planes. On P3 while evidence of slip activities on (101) , (101) and (011) are observed, $(\bar{1}01)$ and (101) planes slip on P4, giving a three-dimensional view of the slipped planes beneath the indented (001) surface.

rotation matrix, $\mathbf{R}$, can be generated as follows, $\mathbf{R} = \mathbf{I} + (\sin \theta)\mathbf{N} + (1 - \cos \theta)\mathbf{N}^2$, where $\mathbf{I}$ is the identity matrix [15]. Using $\mathbf{R}$, the streaking direction of a peak can be identified. Here, the fact that spots streak along multiple directions is indicative of slip occurring on different slip systems in the two adjacent areas of the pillar under the indenter tip. It is known that $\{101\}$ are the primary slip planes in MgO at room temperature, although it can also deform by slip on $\{100\}$ planes – however the stress required to initiate slip on the latter is almost 40 times of that needed on the former [2]. This is shown in the schematic of Fig. 9. Careful observation of the streaked spots from P3 and P4 shows that there are at least two streaking directions associated with each – this renders the curved appearance to the reflections. In P3, the spot streaks to one direction in the top while at the bottom it streaks along two directions with a continuum of intensity between these two extremities. Spot P4 also streaks along one direction in the top and also along one other direction in the bottom. This clearly suggests that two different modes of deformation are operative in these two adjacent regions under the indent. Superimposing, the streaking directions for the $\bar{1}13$ reflection, assuming slip on primary slip planes, the exact $\{110\}$ slip planes responsible for each streaking vector can be estimated. This is shown in Fig. 9. Streaking on the top of P3 corresponds to slip on (101) plane while the two streaking directions at the bottom correspond to simultaneous slip on (011) and (101) planes. On the other hand, in spot P4 the upper and lower streaks are caused by slip on the $(\bar{1}01)$ and (101) planes. In the cubic MgO crystal, these planes have also been schematically represented in Fig. 9. The above observations clearly indicate that slip occurs in the $\{101\}$ planes, which are all at 45° to the loading $[001]$ axis. While on the left of the indent three slip planes are activated, the right hand side of the pillar only has slip on one plane – although it is difficult to dismiss if the streaking due to (101) slip in P3 is overlapped from position P4 due to proximity of the scanning step. From the streaking of the spots, no slip activity was observed on the $\{110\}$ -type planes, which are normal to the sample surface or the loading axis.

The simulations performed also supports the streaking patterns observed in the experiments. The simulated plastic strains for each

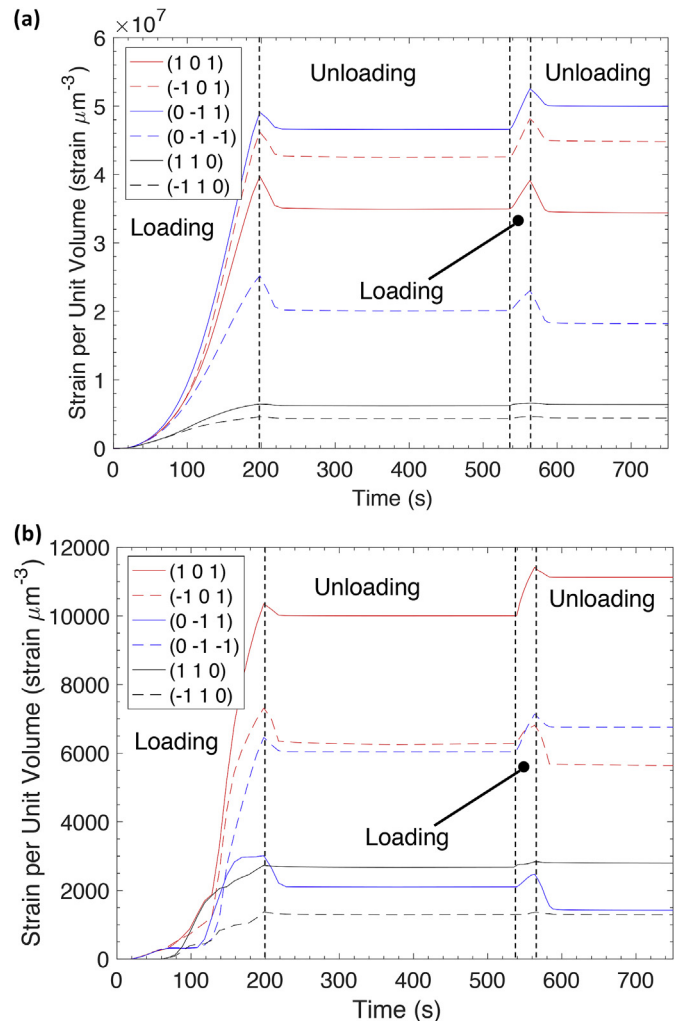


Fig. 10. Plots showing plastic strain on different planes at points (a) P3 (b) P4 obtained from modelling.

slip system per unit volume that correspond to positions P3 and P4 are plotted in Fig. 10. The strains per unit volume (y-axis of the curve) were obtained dividing the total shear strains (obtained from Eq. (6)) by the volume of the individual elements and then summed across the regions of interest shown in Fig. 3, according to Eq. (10):

$$\gamma_{\alpha}^{\text{Per Volume}} = \sum_i \left| \frac{\gamma_{\alpha}^e}{V_e} \right| \quad (10)$$

where N_e is the number of elements and superscript and subscripts e denote element, V_e is the volume of the element and α is the slip system. The strain plots in Fig. 10 show the amount of plastic deformation that occurs in each individual slip system. As seen in Fig. 10, the plastic slip increases as the MgO specimen is indented during each loading cycle and a partial recovery of the plastic strain is clearly evident followed by a period of constant strain during the unloading cycles. This trend is quite consistent across all primary slip systems with exception of the (110) and $(\bar{1}\bar{1}0)$ planes, which stays nearly constant during subsequent periods following the first loading phase. This is consistent with the experimental observations showing no streaking along the {110} planes.

The total amount of plastic strain on the (10 $\bar{1}$), $(\bar{1}01)$ and (0 $\bar{1}\bar{1}$) planes is observed to be significantly higher compared to the (110) and $(\bar{1}\bar{1}0)$ planes at P3 (Fig. 10) suggesting that there is a large amount of plastic deformation occurring in these slip system. This is in accordance with the streaking patterns found in Fig. 9(a) that also suggest large amount of plastic deformation in these planes. Additionally, a part of the plastic strain is recovered during the unloading of the indenter suggesting that there is some recovery of the induced plastic deformation upon unloading, which is also in agreement with the experimental observations of the streaking of the Laue reflection reverting back. Similarly, the cumulative slip for the (10 $\bar{1}$), $(\bar{1}01)$ and (0 $\bar{1}\bar{1}$) planes are also significantly higher compared to that at the other planes as seen in Fig. 10(b), which is consistent with Fig. 9(b), where streaking in the Laue patterns was observed for the (10 $\bar{1}$), $(\bar{1}01)$ and (0 $\bar{1}\bar{1}$) planes – particularly at position P4, the plastic strain on (101) plane obtained from the modelling showed significantly higher levels compared to other slip systems and this is precisely what was observed from the experimental analyses.

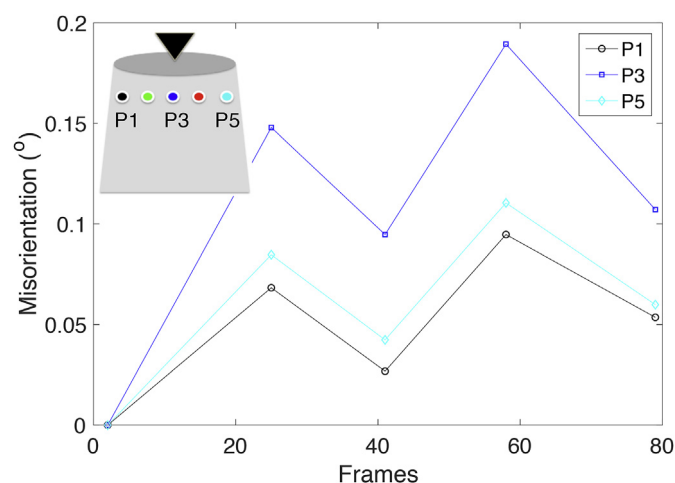


Fig. 11. Plot showing the magnitude of lattice rotation (in degrees) measured from tracking the peak centres of the (113) Laue reflection following Gaussian fitting, with cyclic indentation. This is done for three different locations, P1 (left), P3 (centre) and P5 (right), of the pillar. Time to generate a frame is 10 s.

The intensity profiles of the Laue peaks were fitted by a 2D-Gaussian function using non-linear least square optimization. The description of the fitting equation is given in Appendix 1. It can be observed from Fig. 5 that the Laue spot from a given location within the pillar not only shows streaking but also rotation from the initial position. This rotation, from multiple peaks, gives a direct indication of the magnitude of the elastic lattice rotation that accompanies the deformation in the material around and beneath the indent. Fig. 11 shows the extent of lattice rotation measured from tracking the movement of the (113) Laue peak, with respect to its initial position on the CCD detector with time and two cyclic indentations; it also compares the magnitude of rotation of the peaks from the regions near and away from the indent position. It can be observed that maximum lattice rotation occurs at position P3 directly under the indent and the effect gradually diminishes moving further away. Following the first unloading, some of this rotation is recovered. A maximum rotation of $\sim 0.2^\circ$ occurs at P3 which decreases to $\sim 0.1^\circ$ on either side of the pillar. Also, it can be observed that the rotation is somewhat larger at P5 compared to P1; this also gives an indication of the slight off-centering of the indenter tip on the pillar during the experiment as alluded to previously.

It is to be noted that from the streaking of the peaks, no discernible slip activity was observed on the {110}-type planes, which are at 90° to the sample surface or the indentation axis. Such an array of dislocations on the 90° planes have been observed by Gaillard et al. [35] during nano-indentation of an (001) MgO sample with a spherical indenter. Besides, the formation of loops of screw dislocations on the 45° aligned {101} planes, pile-up of dislocations was also observed on the (110) plane, normal to the indentation surface using a spherical tip. These loops had an edge character. But such pile-ups on the (110) planes have not been reported with a Berkovich indenter [10,11]. This study further confirms that the activation of slip on the 90° planes are non-existent under a Berkovich indenter perhaps due to geometrical considerations.

4.1.3. Evaluation of reversible hysteresis: experimental Laue patterns and finite element modelling

Plastic deformation has been found to initiate in (001)-oriented MgO at loads as low as 0.6 mN, whereby the first dislocations are found to nucleate around the periphery of the indent and significant slip activities and a high density of dislocation loop propagation is observed at 80 mN load [11]. These dislocation loops, with a strong screw character [7], have a high propensity of cross slipping on parallel {110} slip planes [11,35,36]. Besides, the primary {110} $\langle 1\bar{1}0 \rangle$ “soft” slip system, slip in MgO can also occur on “hard” {001} $\langle 110 \rangle$ system, though at larger strength where by slip on the latter system is hindered by considerable lattice resistance [2,37]. While the soft slip system, deforms easily as mentioned previously, the hard slip system is far less compliant. Due to this severe plastic anisotropy, a component of the reversible strain is associated with the elastic relaxation of the hard slip system that has been manifested as reversible hysteresis in cyclic indentation tests up to 6 mN load [12]. A careful look at the time-resolved Laue spots in Fig. 5 shows that streaking of the spots increases with loading while upon unloading a fraction of the streaking is recovered indicating a reversal of plastic flow upon unloading which results in the mechanical hysteresis behaviour.

The hysteresis in the cyclic indentation was also observed in the modelling results in this study, in Fig. 4(c), and can be explained by the plastic anisotropy hypothesis explained previously [12]. Fig. 12 shows the evolution of plastic strains per unit volume for both the primary {101} $\langle 10\bar{1} \rangle$ and secondary {001} $\langle 110 \rangle$ slip systems versus the displacement of the indenter. The plastic strains per unit

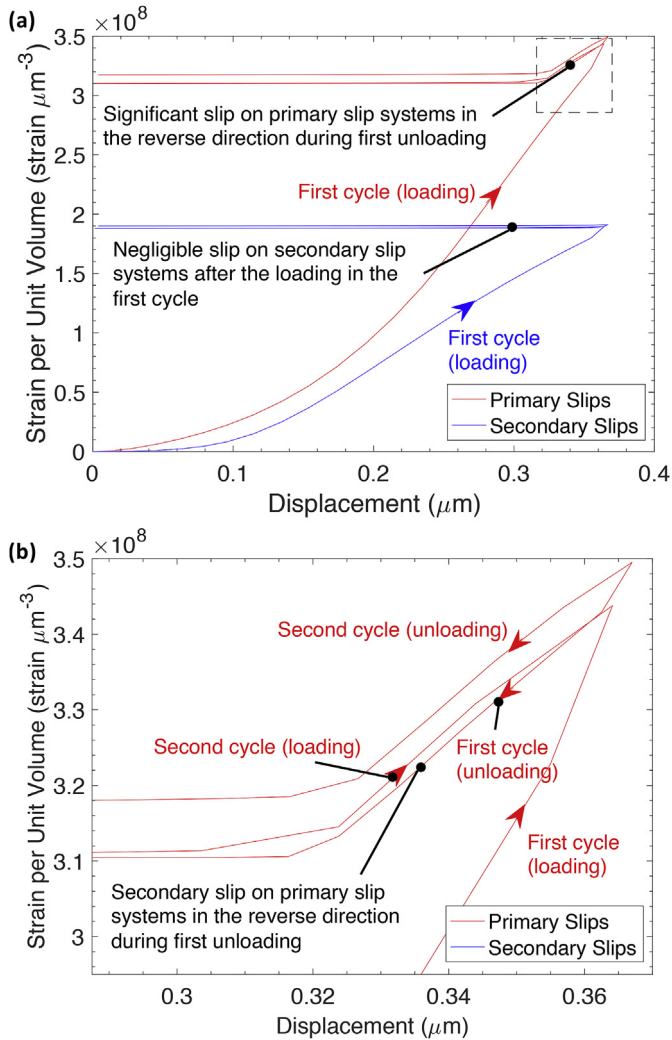


Fig. 12. (a) Cumulative plastic strains per unit volume for the primary and secondary slip systems versus the displacement of the indenter. (b) A magnified section of the square region shown in (a) corresponding to maximum displacement.

volume in Fig. 12 were calculated by normalising the slip divided by the volume of each element for all the elements in the simulation since the elements have different volumes; the strains were added for forward slip on a given slip system during indentation and deducted for reverse slip during unloading. As seen in Fig. 12, the plastic strains for both families of slip systems increases during the loading of the first cycle. However, during the unloading cycle, the secondary slip systems unload elastically. On the primary slip systems, some of the plastic strain is also seen to reverse. This result suggests that during unloading, firstly, the elastic energy stored in the secondary slip systems is released and secondly there is reverse slip simultaneously occurring in the primary slip systems. Then in the second cycle, the secondary slip systems experience negligible slips throughout the second cycle implying that the secondary slip systems deform elastically. On the other hand, the primary slip systems experience slip in both the loading (forward slip) and unloading (reverse slip) of the second cycle. Therefore, in the second cycle, the elastic strains are built up in the secondary slip systems during loading and are released during relaxation when the indenter is unloaded, while the primary slip systems undergoes plastic deformation during loading with reverse during unloading. It is known that microscopic variations in the loading conditions,

stress state, component design or material microstructure, can result in variations in the elastic stress field that can lead to localised plastic deformation below the macroscopic elastic limit. Also, localised elastic deformation can occur above the macroscopic elastic limit due to relative difficulty in activating plastic deformation mechanisms under specific stress states in specific locations within a material system and in this case the high stiffness of the secondary slip systems is the reason for residual elasticity. It is suggested that the plastic recovery in the primary slip system is driven by this stored elastic energy in the hard secondary slip systems and this characteristic behaviour of MgO is manifested in the form of streaking and shrinking of Laue reflections during loading and unloading cycles respectively.

4.2. Indentation cross-section TEM and modelling validation

This differential slip behaviour under the indenter is also shown by the bright field TEM image of the foil sliced out of the MgO pillar, using FIB, in Fig. 13(a–d). The depression from the Berkovich indent is clearly visible in Fig. 13 and could be directly measured from the image to be ~ 350 nm which is in good agreement with the indentation depth (355 nm) assumed in the finite element modelling for the peak load. Looking down the general [100] direction, the zone axis showed two misoriented diffraction patterns from adjacent areas. The diffraction pattern clearly shows streaking and split reflections in Fig. 13(b) obtained by placing the selected area diffraction aperture between the parent and misoriented crystals; this was performed separately on the two misoriented volumes. Selecting the streaked part of the diffracted (020)-reflection in either case, the dark field images were obtained that showed differential illumination of two adjacent deformed sub-volumes under the indent (Fig. 13(c and d)). This clearly shows the development of two misorientated subgrains formed as a result of activation of slip on adjacent areas under the indent. The heavily deformed subgrains were about 500 nm in size. By measuring the degree of misorientation between the spots, an in-plane rotation of $\sim 3^\circ$ was obtained between the substrate and misoriented subgrain – this is comparable to the magnitude of lattice rotation observed by Laue peak tracking, i.e. 2° , which is striking when this is

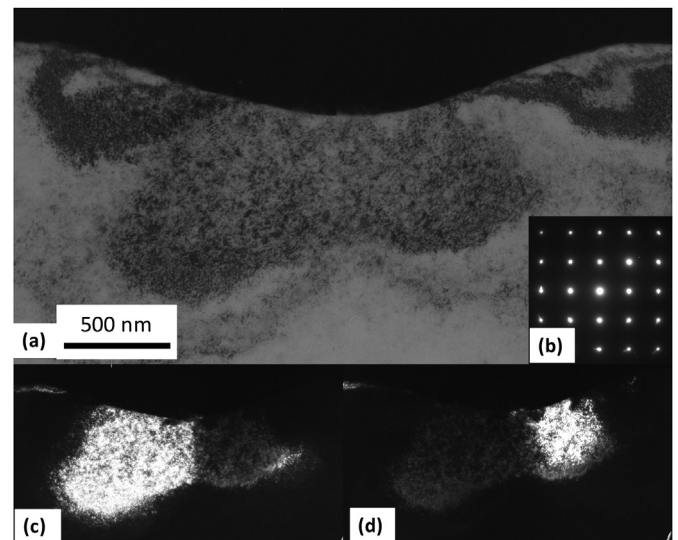


Fig. 13. (a) TEM bright-field micrograph of the area under the indent imaged down the (b) [100]-direction showing the recess on the surface caused by the indenter. (c and d) Dark-field images clearly showing the formation of two distinctly misoriented subgrains formed beneath the indenter.

measured independently from the two experimental techniques.

There is a strong interaction of orthogonal $\{110\}$ slip planes under the indenter in $[001]$ oriented MgO. As the indenter tip is forced into the surface, slip starts occurring on the 45° oriented $\{101\}$ planes. The downward movement of the tip causes further slip by nucleation and subsequent glide of unpaired dislocations along the orthogonal $\{101\}$ planes diverging out from the point of contact on the surface. As $\frac{a}{2} \langle 101 \rangle$ dislocations glide further deep into the sample, along the orthogonal $\{101\}$ planes, they interact to form $a[100]$ dislocations through the reaction $\frac{a}{2}[101] + \frac{a}{2}[\bar{1}0\bar{1}] = a[100]$. Now $a[100]$ dislocations being sessile on $\{101\}$ planes lock the sessile $\frac{a}{2} \langle 101 \rangle$ dislocations. However, in the present experiment, the white beam was scanned at a depth of $\sim 1 \mu\text{m}$ from the top of the pillar. And hence, the only signature borne by the Laue spots was that of simultaneous activation of slip on the $\{110\}$ planes. Out of the 4 possible $\{101\}$ planes aligned at 45° to the $[001]$ axis, the activation of three $\{101\}$ -type planes was observed through the streaking of the Laue spots. Even earlier studies of deformation mechanisms conducted through etch-pit characterisation suggested the nucleation and movement of the first few dislocation loops on $\{101\}$ planes [10]. Slip on one set of $\{110\}$ planes is hindered through slip bands formed on a conjugate $\{101\}$ slip planes, especially for thicker bands [1,3,7]. And this leads to appreciable areas where multiple-slip does not occur and hence resulting in non-uniform internal strain.

Fig. 14 shows the various contour plots of the cumulative slips for the different slips systems. The reference axes for the plots have been chosen to concur with the beam direction in the micro-Laue experiment – this implies that the viewing direction is fixed parallel to the $[100]$ -direction. As seen in Fig. 14(e and f), the slips for the $(1\bar{1}0)$ and (110) planes are significantly lower compared to the other slip systems. This result suggests that the slip systems

corresponding to these planes that are normal to the indentation surface are not activated, which is consistent with the experimental observations. On the other hand, as seen in Fig. 14(a and b), the slips in the (101) and $(\bar{1}01)$ planes are high suggesting that these slip systems are the main source of plastic deformation in the indentation process. However, the slips in these planes appear to be symmetrical. This corroborates the experimental observation of the slip on the (101) plane on either sides of the indenter in positions P3 and P4. However, experimental results showed $(10\bar{1})$ slip only at P3 and not at P4 – this might be either due to the misalignment in the viewing direction from the exact $[100]$ in the experiment or inadequate exposure time during scanning of pillar leading to a lower definition of streaking.

The slips in the (011) and $(0\bar{1}1)$ planes in Fig. 14(c and d) were also found to be high. Again, this result shows that slip on these planes are also significant plastic deformation mechanisms in the indentation process. However, unlike the (101) and $(\bar{1}01)$ planes, the (011) and $(0\bar{1}1)$ planes are not symmetrical. In Fig. 14(c and d) it can be seen that larger slip occurs on the left and right sides of the specimen. This is in agreement with the Laue results, which showed slip on (011) plane only at position P3. Simulation results suggest that there is a mutual misalignment between the left and right side of the crystal and this is also confirmed by the TEM results shown in Fig. 13. Furthermore, slip occurs on opposite sides of the crystal structure for the $(0\bar{1}1)$ and (011) slip systems. This result indicates that the crystal slips in different directions in the two regions, which leads to the formation of the two misoriented subgrains.

4.3. Elastic deformation gradient under the indenter

Persistent slip within a (constrained) sample gives rise to the

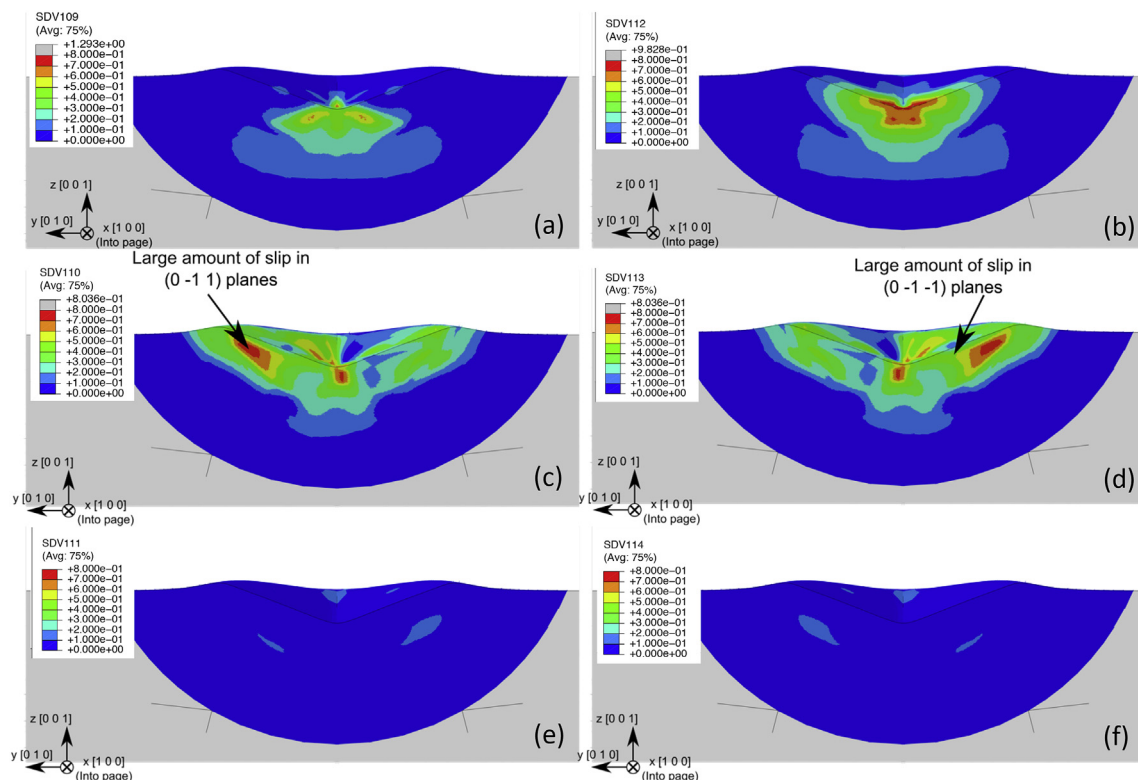


Fig. 14. Contour plots of cumulative slips after final unloading (a) (101) (b) $(\bar{1}01)$ (c) $(0\bar{1}1)$ (d) (011) (e) $(1\bar{1}0)$ (f) (110) .

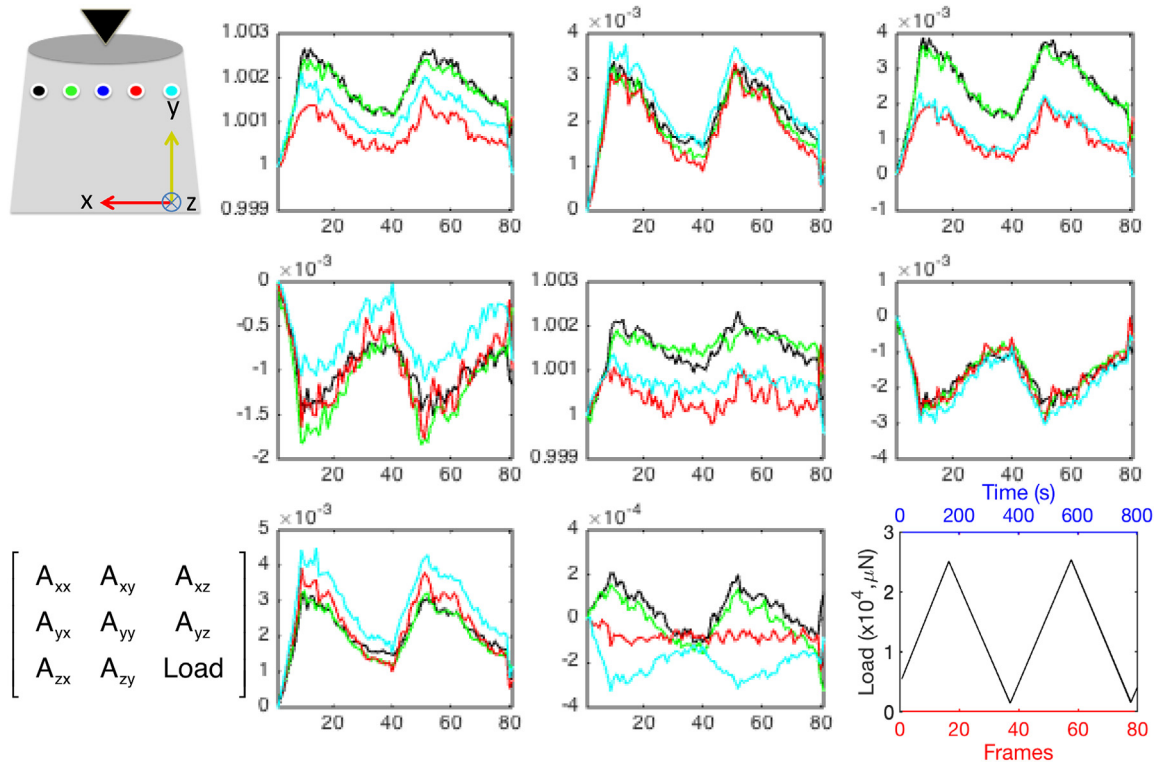


Fig. 15. The development of elastic displacement field beneath the indenter obtained from different locations of the pillar.

development of short-range deviatoric strain fields within the sample volume, the components of which are captured appreciably by Laue diffraction. To calculate the deformation gradient, for a given location on the micropillar, the peak positions (i.e. the peak-centre coordinates on the detector) were tracked with time. This was done by fitting the peaks with a 2D-Gaussian function, as outlined in Appendix 1, in order to obtain the coordinates for the centres of the peak for every frame. For this a minimum of four reflections had to be chosen so as to uniquely obtain the deformation gradient and in this case six reflections were followed with time. These peaks were fitted simultaneously and their positions recorded. Once the peak-centres were identified, the complete three-dimensional diffraction vectors were drawn. The details of this procedure are given in Appendix 2. By minimizing the differences between the final and initial diffraction vectors, the components of the deformation gradient tensor could be mapped for every frame of time. And applying the same calculations for the different rastered positions across the width of the micropillar, a spatial map of the deformation gradient, F_{ij} , was also obtained. Thus using the deformation tensor, the final position of a vector, $\vec{n}'$, within the sample can be obtained from the initial vector, $\vec{n}$, as

$$\vec{n}' = F_{ij} \cdot \vec{n} \quad (11)$$

Now corresponding displacement gradient, A_{ij} , could be obtained from Eq. (12):

$$F_{ij} = I + A_{ij} \quad (12)$$

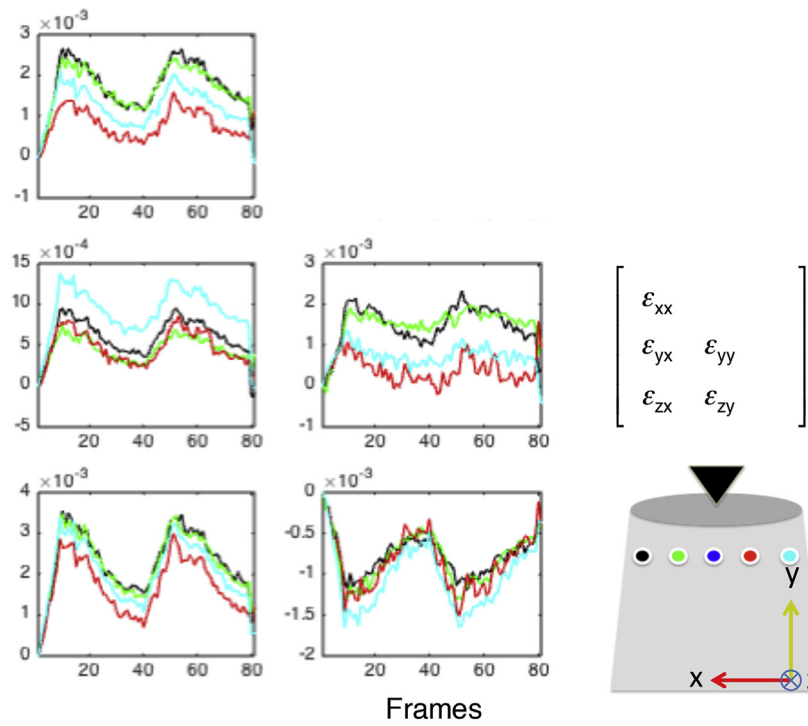
where I is identity matrix. For a given vector $U = [U_x, U_y, U_z]$, the displacement gradient is defined as

$$A_{ij} = \begin{bmatrix} \frac{\partial U_x}{\partial x} & \frac{\partial U_x}{\partial y} & \frac{\partial U_x}{\partial z} \\ \frac{\partial U_y}{\partial x} & \frac{\partial U_y}{\partial y} & \frac{\partial U_y}{\partial z} \\ \frac{\partial U_z}{\partial x} & \frac{\partial U_z}{\partial y} & \frac{\partial U_z}{\partial z} \end{bmatrix} = \vec{\epsilon} + \vec{\omega} \quad (13)$$

where $\vec{\epsilon}$ and $\vec{\omega}$ are the strain and rotational components of the gradient.

The evolution of the displacement tensors at different locations on the pillar is shown in Fig. 15. In Laue diffraction, the position and energy of the Laue spots is dependent on the orientation of the crystal and lattice parameters but generally the energies of the diffracted spots are not measured and so white beam Laue diffraction is usually insensitive to hydrostatic strain. In general, the variation of the elastic displacement gradient closely follows the profile of the loading cycle. The two components, strain and rotations, can be resolved from the displacement gradient. The temporal evolution of these components from areas on either sides of the indent is mapped in Fig. 16. Fig. 16(a) shows that the elastic strains show a similar trend of variation across the pillar with only the ϵ_{zy} component showing compressive strains. Some variation in the ϵ_{yy} -component along the principle y-direction is observed – with the strain values somewhat higher on one side (left) of the indent (represented by P1, P2) as compared to the other (P4 and P5), which is attributed to the off-centred indent position with respect to the pillar. The components of the rotation tensors, Fig. 16(b) also show a similar overriding trend, along the width of the pillar, except for the ω_{zx} -components, which indicate positive rotations on the right side of the indent (P4 and P5) compared to

(a)



(b)

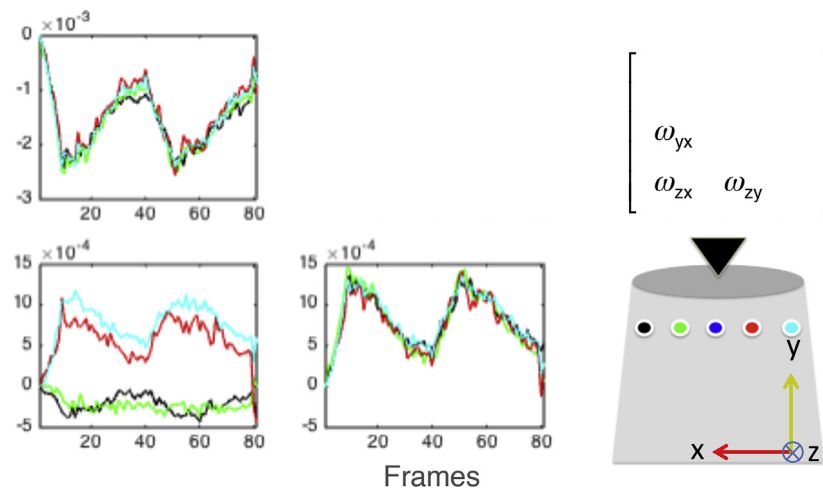


Fig. 16. The temporal evolution of the elastic components, (a) deviatoric strain and (b) rotation, from areas on either sides of the indent. Duration of each frame is 10 s.

negative on the left (P1 and P2). Such opposite lattice rotations are expected to occur on either sides of the indenter in order to accommodate the geometric constraints of the indenter. It is believed that a differential behaviour in these elastic components plays an important role in creating the final misorientated sub-grain upon continued deformation. Through controlled loading and micro-Laue diffraction experiments, it is demonstrated that detailed time-resolved quantitative information on the elastic behaviour of a sample could be obtained which otherwise is not so trivial to acquire through other experimental protocols.

5. Conclusion

This article undertakes a detailed *in-situ* study coupled with micro-Laue diffraction and nano-indentation to investigate the progress of deformation mechanism in [001]-oriented single crystal MgO. By scanning the pillar across with a micro-focussed beam, both time and spatially resolved mapping of deformation during nano-indentation with a peak load of 25 mN was performed. The following findings were obtained through the study.

- Upon indentation, the Laue spots showed both rotation and streaking which are indicative of both elastic lattice rotation and induced plastic strain in the material respectively. Multiple facets of streaking of the Laue peaks suggested plastic slip occurring on almost all the {101}-type slip planes oriented 45° to the sample loading direction with, however, no indication of slip on the 90° {110} planes. Crystal plasticity finite element modelling of [001]-MgO under the indent also showed the least contribution of slip from the planes normal to the indentation surface for a Berkovich indenter. The crystal plasticity modelling results also clearly showed occurrence of slip on the {101}-type planes as indicated by the Laue results – a good agreement between the experimental and simulated slip distribution under the indent on the 45° planes was also observed.
- Owing to separate sets of conjugate {101} slip systems operating on either sides of the pillar beneath the indenter, sub-grains were found to nucleate with distinct misorientation between them, which is also corroborated by post-mortem TEM studies.
- Through cyclic loading the phenomenon of hysteresis in MgO is clearly revealed by the expansion and contraction of the Laue reflections driven by reversal of plastic strain on {110} planes that is in turn caused by the stored elastic energy in the hard {100} secondary system. The magnitude of the plastic strain recovery in the primary slip system has been obtained in the crystal plasticity simulations.
- By tracking the peak movement, it was observed that greater degree of lattice rotation occurred in the material under the indent, measured to be 0.2° as compared to far field. With the progress of deformation, full field quantitative elastic strain and rotation gradients could be constructed. While the elastic strains showed a similar overriding variation across the pillar with applied stress, opposite rotational (ω_{zx}) gradients were clearly observed on two sides of the micropillar under the indent, which is the precursor to the final sub-grain formation.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.actamat.2017.12.002>.

Appendix 1

The intensity of the Laue peaks (I_{calc}) on the detector were fitted using a 2D-Gaussian function in MATLAB as follows

$$I_{calc} = A * \exp(-a(x - x_c)^2 + 2b(x - x_c)(y - y_c) + c(y - y_c)^2), \quad (A1)$$

where

$$a = \frac{\cos^2 \theta}{2s_1^2} + \frac{\sin^2 \theta}{2s_2^2}$$

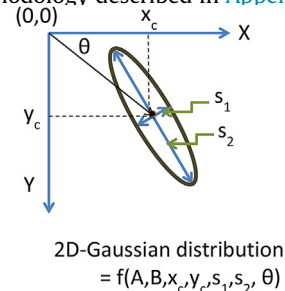
$$b = -\frac{\sin 2\theta}{4s_1^2} + \frac{\sin 2\theta}{4s_2^2}$$

$$c = \frac{\sin^2 \theta}{2s_1^2} + \frac{\cos^2 \theta}{2s_2^2}$$

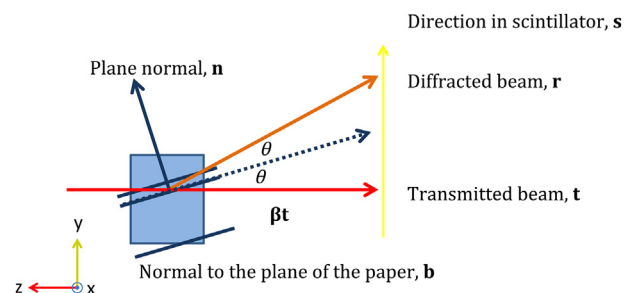
Here A and B are the peak and background intensities, x_c and y_c are the centre coordinates of the peak, s_1 and s_2 are the major and minor axes of an elliptical peak, and θ is the angle of inclination (clockwise) of the peak with the x-axis. All the seven parameters were refined to obtained a least square fit of the measured (I_{meas}) and calculated (I_{calc}) intensity as

$$y = \sum_i (I_{calc} - I_{meas})^2 \quad (A2)$$

This non-linear least square fitting was done by using the optimization toolbox of MATLAB. The fitting routine was followed for 6 peaks for all every frame. Once the centres of the spots are obtained in the detector coordinates from given the actual sample-detector distance, the diffraction vector for each peak can be obtained by the methodology described in Appendix 2.



Appendix 2



Let the sample plane normal vector for a given set of (hkl) plane be

$$\vec{n} = \begin{bmatrix} \vec{n}_x & \vec{n}_y & \vec{n}_z \end{bmatrix} \quad (A3)$$

where $\vec{n}_x^2 + \vec{n}_y^2 + \vec{n}_z^2 = 1$ and

$$n = ha^* + kb^* + lc^* \quad (A4)$$

where a^* , b^* and c^* are the reciprocal lattice vectors in the given crystal system.

According to the selected coordinate system,

$$\vec{t} = [0 \ 0 \ -1] \quad (A5)$$

The direction within the sample, normal to the plane of the paper is evidently given by the cross product of $\vec{t}$ and $\vec{n}$, as seen from the figure

$$b = \vec{t} \times \vec{n} = \begin{bmatrix} \vec{n}_y & -\vec{n}_x & 0 \end{bmatrix} = \vec{b} \sin(90^\circ + \theta) = \vec{b} \cos(\theta) \quad (A6)$$

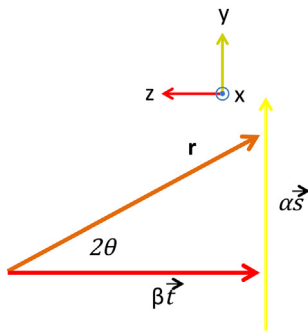
Therefore, it follows

$$\cos \theta = \sqrt{\vec{n}_y^2 + \vec{n}_x^2} \quad (A7)$$

The dot product of $\vec{t}$ and $\vec{n}$ is given as

$$\begin{aligned} \frac{\vec{r}}{\alpha} &= \begin{bmatrix} \frac{\vec{n}_x}{\sqrt{\vec{n}_y^2 + \vec{n}_x^2}} & \frac{\vec{n}_y}{\sqrt{\vec{n}_y^2 + \vec{n}_x^2}} & 0 \end{bmatrix} + \begin{bmatrix} 0 & 0 & -\frac{1 - 2\vec{n}_z^2}{2\vec{n}_z\sqrt{\vec{n}_x^2 + \vec{n}_y^2}} \end{bmatrix} \\ \frac{\vec{r}}{\alpha} &= ar = \frac{1}{\sqrt{\vec{n}_y^2 + \vec{n}_x^2}} \begin{bmatrix} \vec{n}_x & \vec{n}_y & -\frac{1 - 2\vec{n}_z^2}{2\vec{n}_z} \end{bmatrix} \end{aligned} \quad (A14)$$

$$\begin{aligned} \vec{t} \cdot \vec{n} &= -\vec{n}_z = \cos(90^\circ + \theta) = -\sin(\theta) \\ \sin \theta &= \frac{\vec{n}_z}{\vec{n}} \end{aligned} \quad (A8)$$



From the above construction of the transmitted beam vector, the diffracted beam and the vertical axis of the detector we can write

$$\begin{aligned} \vec{r} &= \alpha \vec{s} + \beta \vec{t} \\ \frac{\vec{r}}{\alpha} &= \vec{s} + \frac{\beta}{\alpha} \vec{t} \end{aligned} \quad (A9)$$

Also from the figure,

$$\cot 2\theta = \frac{\beta}{\alpha} = \frac{\cos^2 \theta - \sin^2 \theta}{2 \sin \theta \cos \theta} \quad (A10)$$

Substituting the $\cos \theta$ and $\sin \theta$ in terms of components of $\vec{n}$ from Equations (A7) and (A8) we have

$$\frac{\cos^2 \theta - \sin^2 \theta}{2 \sin \theta \cos \theta} = \frac{\vec{n}_y^2 + \vec{n}_x^2 - \vec{n}_z^2}{2\vec{n}_z\sqrt{\vec{n}_x^2 + \vec{n}_y^2}} = \frac{1 - 2\vec{n}_z^2}{2\vec{n}_z\sqrt{\vec{n}_x^2 + \vec{n}_y^2}} = \frac{\beta}{\alpha} \quad (A11)$$

The direction of the detector, s , can be expressed as the cross product of the normal to the plane of the paper, b and the transmitted beam vector, t

$$s = b \times \vec{t} = \begin{bmatrix} \vec{n}_x & \vec{n}_y & 0 \end{bmatrix} \quad (A12)$$

Expressing in terms of unit vector

$$\vec{s} = \frac{1}{\sqrt{\vec{n}_y^2 + \vec{n}_x^2}} \begin{bmatrix} \vec{n}_x & \vec{n}_y & 0 \end{bmatrix} \quad (A13)$$

Using Equations (A11) and (A13), Equation (A9) can be rewritten as

Replacing $= \frac{1}{a'}$, the magnitude of the diffraction vector in Equation (A14) is given by $|a'r| = \frac{1}{2\vec{n}_z\sqrt{\vec{n}_y^2 + \vec{n}_x^2}}$

Hence the unit diffraction vector is

$$\begin{aligned} \vec{r} &= \frac{\frac{1}{\sqrt{\vec{n}_y^2 + \vec{n}_x^2}} \begin{bmatrix} \vec{n}_x & \vec{n}_y & -\frac{1 - 2\vec{n}_z^2}{2\vec{n}_z} \end{bmatrix}}{\frac{1}{2\vec{n}_z\sqrt{\vec{n}_y^2 + \vec{n}_x^2}}} \\ &= 2\vec{n}_z \begin{bmatrix} \vec{n}_x & \vec{n}_y & -\frac{1 - 2\vec{n}_z^2}{2\vec{n}_z} \end{bmatrix} \end{aligned}$$

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