In-situ, radial X-ray diffraction study of texture and stress during phase transformations in bcc-, fcc-, and hcp-iron up to 36 GPa and 1000 K

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Abstract

Using a newly developed experimental protocol, based on externally heated diamond anvil cells, in situ stress and textures studies are performed on polycrystalline bcc- (α -), fcc- (γ -), and hcp- (ϵ -)Fe during plastic deformation up to 36 GPa and 1000 K. At 7 GPa and 973 K, α -Fe displays a texture attributed to dominant slip on $\{110\}\langle\bar{1}11\rangle$. Upon pressure increase up to 24 GPa at constant temperature, differential stress is shown to increase to 0.6 ± 0.1 GPa while Fe is transforming into γ -Fe and ϵ -Fe. Textures observed in γ -Fe are fully induced by phase transformation with little effects of plastic deformation. Textures observed in ϵ -Fe are the result of phase transformation from γ -Fe combined with plastic deformation with dominant basal $(0001)\langle 11\bar{2}0 \rangle$ slip. The differential stress measured in ϵ -Fe is consistent with a shear strength of ~ 1.9 GPa at 17 GPa and 300 K, with a temperature and pressure dependence of $-0.003 \text{ GPa.K}^{-1}$ and 0.074, respectively.

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1 Introduction

Phase transformations in metals attract a large interest because of their implications to understand materials behavior, shape memory alloys, and the fundamental physical processes driving phase transitions. In particular, phase transformations can affect the texture as crystallographic relationships and variant selection can occur between the parent and product phases [1]. However, most experiments regarding textures and phase transformations are performed at ambient pressure and very little is known regarding phase transitions at pressure above 1 GPa. Similarly, strength is a fundamental property for constraining materials behavior [2] and the rheology of deep Earth minerals [3]. As such, measurement of materials strength at high pressure and high temperature is key to understanding materials behavior at these conditions.

Recently, we have developed a new diamond anvil cell (DAC) technique allowing the study of stress and texture in polycrystals, in-situ, up to pressures and temperatures of 50 GPa and 1500 K [4]. Iron is a good candidate for testing the capabilities of this experimental technique due to its well characterized high pressure – high temperature phase diagram (Fig. 1) and its importance to engineering, fundamental physics, and deep Earth geodynamics. Its plastic properties have also been well studied under high pressure [5,6,7,8,9,10,11]. At ambient conditions iron takes a bcc (α) structure and, upon heating, transforms to the fcc structured γ phase. This transformation in steel is of critical importance in engineering because there is a significant reduction in materials strength associated with the transformation. The high pressure ϵ phase has an hcp structure and is believed to be the main constituent of the Earth's inner core [12].

Based on considerations on crystal structures and microscopic observations, several orientation relationships for the bcc-fcc phase transformation have been proposed in the literature [13,14] (Table 1), including those of Bain [15], Kurdjumov–Sachs [16], and Nishiyama-Wassermann [17], but they have rarely been studied in-situ under high pressure and/or high temperature [18]. Similarly, the bcc to hcp transformation in Fe has been extensively studied and modeled [19,20,21,22] and most proposed mechanism lead to orientation relationships resembling those proposed by Burgers and originally identified in zirconium [23]. Finally, orientation relationships, sometimes labeled as Shoji-Nishiyama, have also been reported for the fcc to hcp phase transformation [1,24,25].

In this paper, we present our new experimental approach and show how it can be used for the direct and in-situ investigation of strength and transitions mechanisms in Fe over a large portion of its phase diagram.

2 Experiment and data processing

2.1 High pressure/ high temperature deformation and in-situ X-ray diffraction

The in-situ radial X-ray diffraction (rXRD) and DAC deformation experiment was performed at beamline 16-BMD of the High-Pressure Collaboration Access Team (HP-CAT), Advanced Photon Source (APS), Argonne National Laboratory and has been extensively described before [4]. Therefore, we will only provide a summary of the main relevant features.

The wavelength of the $15 \times 15 \mu\text{m}^2$ monochromatic x-ray beam was tuned to $0.38745 \text{ \AA}$. Collection time for diffraction images was 15 minutes on a 3450×3450 pixel Mar345 online image plate. Sample to detector distance, detector tilt, and pixel size ratios were calibrated using a CeO_2 standard (NIST 674b). The sample, $20 \mu\text{m}$ in thickness and $80 \mu\text{m}$ in diameter,

was a pure commercial powder of iron (Alfa Aesar, 99.9 % purity) with a starting grain size of 1-2 μm . A small piece of 5 μm thick gold foil was added for additional pressure calibration but could not be seen on the rXRD images. Both were loaded in a high temperature, resistive heating DAC designed for rXRD equipped with 300 μm tip diameter flat anvils and a gasket consisting of an amorphous boron and epoxy ring confined with a sheet of mica [4]. Heating of the sample is achieved by using 1 mm thick graphite heaters that surround the sample assembly and the tips of the diamonds. Deformation is achieved by increasing the pressure load through a membrane attached to the back of the DAC. Temperature is measured with two Pt-Pt13Rh (type R) thermocouples with an accuracy of the thermocouple reading of ± 5 K. Based on differences between the reading on both thermocouples, we estimate that our uncertainty on temperature and temperature gradients do not exceed 60 K. Pressure is estimated from the diffraction of the sample itself using a previously calibrated equation of state (EoS) (see below). The overall experiment was performed over 37 hours (Table S1).

2.2 Data processing

Diffraction images were quantitatively analyzed for cell parameters, phase proportions, texture, grain sizes, and lattice strains using the Rietveld method as implemented in the software package MAUD [26] according to the procedure described previously [8]. Representative refinement results are shown in Fig. 2 for γ - and ϵ -Fe at 31 GPa and 900 K. Diffraction images were integrated over 5° increments of the azimuth angle, resulting in 72 slices. Data were fit using a bcc, fcc, and hcp structure with adjustable phase ratios. The variations in peak positions with azimuth were adjusted assuming the lattice strain equation of Singh et al [27]. For the determination of crystallite size, Popa line broadening with an isotropic size-strain model was used [28] while the instrument broadening had been calibrated with the CeO_2 standard.

Pressures are calibrated using X-ray diffraction of an internal standard, the EoS of the sample itself, iron, in the bcc structure [29], and later hcp structure [30], to estimate pressure at any given temperature during the experiment. To correct for the effect of non-hydrostatic stresses, pressures were calculated using the hydrostatic cell parameters of α - and ϵ -Fe as derived from the MAUD refinements. The temperature-pressure path explored during the experiment is shown in Fig. 1.

Differential stress was calculated using the model of Singh et al [27] and the aggregate shear modulus of each phase. For α -Fe, it was measured under high pressure and ambient temperature [31] and under high temperature of ambient pressure [32]. Therefore, we used a simple model assuming a linear combination of both measurements. The shear moduli of ϵ - and γ -Fe are not well constrained under combined high pressure and high temperature. For ϵ -Fe, ambient temperature ultrasonic measurements at 16.5 GPa [33] indicate that $K/G = 2.75$ while a combination of inelastic X-ray scattering and X-ray diffraction leads to $K/G = 2.62$ at 52 GPa [34]. We therefore assumed $K/G = 2.7$ for ϵ -Fe calculating the aggregate bulk modulus at pressure and temperature using the EoS of ref. [30]. There are still significant debates regarding the EoS of γ -Fe (e.g. [29]). Thus, we took an even more simplified approach, assuming that the shear modulus of γ -Fe was equal to that of α -Fe. Phase fractions, grain sizes and stresses deduced at each measured point are shown in Fig. 3. Times, pressures, temperatures, phase fractions, cell parameters, grain sizes, and stresses are also listed in Table S1.

Lattice preferred orientations (LPOs) were fit using the E-WIMW algorithm with an orientation distribution function (ODF) resolution of 7.5° and assuming cylindrical symmetry about the compression direction. The ODF was further smoothed with a 10° Gauss filter in

BEARTEX [35]. Axial compression textures can be compactly represented by an inverse pole figure (IPF), showing the relationship between crystallographic directions of crystallites to the compression direction. Fe has cubic or hexagonal crystal symmetry; thus, only portions and the inverse pole figure are needed to represent the ODF. Pole densities are given in multiples of random distribution (mrd), where mrd = 1 corresponds to a random distribution and an mrd of infinity corresponds to a single crystal texture.

Refinements for the bcc phase were based on measurements of the 110, 200, and 211 diffraction lines. Refinements for the fcc phase were based on measurements of the 111, 200, and 220 diffraction lines. Refinements for the hcp phase were based on measurements of the $10\bar{1}0$, 0002, $10\bar{1}1$, $10\bar{1}2$, and $11\bar{2}0$ diffraction lines. Textures measured in all three phases are summarized in Fig. 4.

3 Results

3.1 Phase diagram and phase proportions

Fig. 1 presents the pressure-temperature path explored during the experiment while the phase proportions of α -, γ - and ε -Fe extracted from the MAUD refinements are shown in Fig. 3. The start of phase transformation between α -Fe and γ -Fe is induced at 7 GPa and 973 K. Pressure is then increased up to 24 GPa at ~980 K, with a gradual transformation of α -Fe into γ -Fe. At 14.5 GPa, α -Fe completely disappears and, increasing pressure above 14.5 GPa, we observe the gradual transformation of γ -Fe into ε -Fe. Between 24 and 33 GPa, temperature is reduced to 900 K and, later, further reduced to 300 K while increasing pressure to 36 GPa. This process results in further transformation of γ -Fe into ε -Fe but a full transition to ε -Fe was never achieved.

3.2 Stress

Differential stresses measured in the three phases of Fe are presented in Fig. 3. First, there does not seem to be any significant difference between stresses measured in all three phases. When reaching the transition to the γ phase at 7 GPa and 973 K, stresses are low, on the order of 0.1 ± 0.1 GPa. When pressure is increased to 24 GPa at 980 K, stress in the sample is 0.6 ± 0.1 GPa. Pressure is then increased to 33 GPa while lowering the temperature to 900 K. During this process, axial strain is imposed on the sample and stresses continuously increases to 1.3 ± 0.1 GPa. At the end of the experiment, temperature is reduced to 300 K at 36 GPa, resulting in an increase of the differential stress to about 3.0 ± 0.4 GPa.

3.3 Lattice preferred orientations

Upon compression, α -Fe develops a texture characterized by maxima in the 001 (1.1 mrd) and 111 (2.1 mrd) directions, a minimum at 011 (Fig. 4). At 7 GPa and 973 K, γ -Fe appears with a texture characterized by a broad and weak maximum ranging between 011 and 014 with minima at 001 and 111. Later, γ -Fe develops a weak texture with a maximum at 011 (1.4 mrd) and minima at 001 and 111. This is associated with a weakening of texture in α -Fe, with a maximum pole density of 1.8 mrd at 111 at image 62. At 16 GPa and 970 K, α -Fe completely disappears. This is correlated with the appearance of a small proportion of ϵ -Fe with a marked maximum at 0001 (2.25 mrd). Upon further transformation and compression, ϵ -Fe develops a strong texture with a maximum at about 30° from 0001 with maximum pole density of 2.11 mrd and a distinct minimum at $10\bar{1}0$.

158 **4 Discussion**

159 **4.1 Phase diagram**

160 Overall, there is a good agreement between our measurements and phase diagrams published
161 previously [30,36]. Full phase transition to ϵ -Fe was never achieved. Indeed it has been
162 reported that kinetics of phase transitions involving γ -Fe are slow and difficult to study
163 experimentally [36,37]. Moreover, stress and pressure gradients are known to affect the
164 pressure range of phase coexistence in Fe. Achieving a full pressure induced phase transition
165 is highly dependent on stress and involves significant hysteresis [38].

166 **4.2 Stress**

167 Very few stress measurements have been performed on Fe polycrystals in the current pressure
168 and temperature range. In axial compression deformation experiments up to 10% axial strain in
169 a D-DIA at 17 GPa, it was found that stresses in ϵ -Fe polycrystals could reach 1.6 GPa at
170 600 K and 1.8 GPa at 400 K [7]. At 300 K, rXRD measurements in the DAC found stresses in
171 the order of 3.0 ± 0.2 GPa for pressures ranging between 15 and 30 GPa [6] and
172 4.5 ± 0.2 GPa at 52 GPa [10]. Those are all very consistent (Fig. 5) with our measurements
173 and indicate that the strength of ϵ -Fe is ~ 1.9 GPa at 17 GPa and 300 K, with a temperature
174 and pressure dependence of $-0.003 \text{ GPa.K}^{-1}$ and 0.074 , respectively.

175 **4.3 Textures in α -Fe**

176 Our measured textures in α -Fe agree with compression textures reported for bcc metals [39]
177 and are attributed to dominant slip on $\{110\}\bar{1}11$. This texture is also consistent with
178 previous texture measurements on α -Fe under non-hydrostatic stress in the DAC [20,40]. At

the start of the experiment, control of the sample pressure was lost while starting the temperature increase. This resulted in a transition into ε -Fe for a brief moment (Figs. 1 and 3). However, this incursion into the ε -Fe stability field does not seem to have influenced our measurement and any effect should be negligible.

4.4 Textures in γ -Fe

After transformation, γ -Fe shows a maximum at 011 and minima at 001 and 111. Transformation textures for the bcc to fcc transformation in steel have been extensively studied and several orientation relationships for the bcc-fcc phase transformation have been proposed in the literature [13,14], including those of Bain, Kurdjumov–Sachs, and Nishiyama-Wasserman (Table 1). The 011 maximum in the fcc texture can also be generated by slip on $\{111\}\langle 1\bar{1}0\rangle$ and is the typical compression texture in fcc metals [39]. It has also been observed in DAC deformation experiments on fcc Cu [41].

In order to test whether our observed texture arise from transformation or plastic deformation, we applied a transformation model based on the Nishiyama-Wasserman orientation relationships with no variant selection (Fig. 6) using the procedure described in Ref. [18]. The obtained textures are in very good agreement with experimental measurements at 16.4 GPa, after the full disappearance of α -Fe (Fig. 4). Moreover, observed textures in γ -Fe do not evolve upon further compression. We therefore conclude that the measured textures in γ -Fe mostly result from the phase transformation and that γ -Fe was only weakly affected by plastic deformation in the experiment. Our measured textures are weak. Therefore, subtle differences between transformation mechanisms cannot be resolved and other orientation relationships such as Bain or Kurdjumov–Sachs would also fit our data.

4.5 Textures in ϵ -Fe

After transformation from γ -Fe, texture in ϵ -Fe shows a strong maximum at 0001 that gradually transforms towards a girdle 30° away from 0001 with a pronounced minimum at $10\bar{1}0$. Unlike previous studies where transition occurred from the bcc phase [20,40], we do not observe a maximum at $11\bar{2}0$ after the phase transition, indicating a different transition mechanism. The observed girdle 30° away from 0001 is different from the pure 0001 maximum observed at 300 K in the DAC [20] and up to 600 K in the D-DIA [7]. On the other hand, it has been observed in samples undergoing laser heating at 1950 K [40]. Therefore it is possible that a different mechanism is active at higher temperatures with a transition temperature ranging between 600 and 900 K.

The observed textures in ϵ -Fe could originate from a transformation from bcc to hcp, fcc to hcp, or from plastic deformation. In order to differentiate those effects, we performed simulations of transition textures that would be obtained from the bcc phase, according to Burgers orientation relationships [23], or from the fcc phase, according to the Shoji-Nishiyama orientation relationships [1,24,25] (Table 1). Results are presented in Fig. 6. In both cases, the transformation textures show a maximum at $11\bar{2}0$ with a secondary maximum 30° away from 0001 and look significantly different from the experimental results. We can therefore conclude that textures in ϵ -Fe are significantly affected by an orientation process, such as variant selection, nucleation, or plastic deformation.

Investigation of variant selection requires data in which the effect of plastic deformation can be easily decorrelated, which is not possible with the present data. Similarly, nucleation under stress is a very plausible mechanism for high temperature phase transformation such as the one between γ - and ϵ -Fe, but our dataset does not allow us to investigate such models.

Therefore, we decided to focus on the effect of plastic deformation and turn to polycrystal plasticity modeling. In those models, texture development depends on the deformation geometry as well as the relative activities of the different deformation modes such as slip or deformation twinning. We used the Los Alamos viscoplastic self-consistent code (VPSC, version 7) [42] that treats each grain as an inclusion in a homogeneous but anisotropic medium that has the average properties of the polycrystal. As deformation proceeds, crystals deform and rotate to generate preferred orientation. By applying different critical resolved shear stresses (CRSSs) to slip systems and twin modes, the model will favor one deformation mode over another. This results in different textures for different combinations of deformation modes.

As a starting texture for ϵ -Fe, we used 2000 grains weighted to resemble that obtained after phase transformation from γ -Fe (Fig. 6). The plastic properties of ϵ -Fe have been extensively studied under high pressure [5,7,9,11,20,40]. Among those, very few apply to ϵ -Fe at high temperature and we selected plastic models that generate textures closely resembling that of Fig. 3, with a maximum approximately 30° away from 0001 (Table 2). In order to match the intensity of the textures observed experimentally at the end of the experiment, we find that an incremental deformation path of 10% compressive strain is appropriate. Texture results are shown in Fig. 6 and values for CRSS and activities of the deformation modes are given in Table 2.

Both models provide textures that broadly agree with the experimental results (Figs. 4, 6). In both cases, deformation is strongly dominated by basal slip (61% and 99% of the total plastic activity for models 1 and 2, respectively). The rest of the deformation is accommodated by prismatic, pyramidal $\langle a \rangle$, and pyramidal $\langle c+a \rangle$ slips and twinning. It is important to note that, however, for both models the activity of tensile twinning is low. Twinning is apparently

suppressed at the high temperatures as observed for Hf [5]. Indeed, in hcp metals with a low c/a ratio, tensile twinning produce a texture with c-axes nearly parallel to the compression direction whereas a combination of basal, prismatic, and pyramidal slip produce a texture with a maximum at about 30° away from the compression direction, as observed in the experiment.

4.6 High pressure high temperature phase transformations in iron

Phase transformations in pure Fe are fundamental as Fe is a model material for numerical calculations [21,43], static high pressure experiments [20,30], shock-wave physics [22], understanding the physical properties of industry relevant materials such as transformation induced plasticity (TRIP)-assisted steels [44], and because the Earth core is believed to be composed mainly of Fe [12].

Our results show that transformation textures between α - and γ -Fe at ~970 K and pressures ranging between 7 and 14.5 GPa are in agreement with a displacive mechanism of the phase transformation and can be modeled using classic orientation relationships (Fig. 4). Transformation textures between γ - and ϵ -Fe above 14.5 GPa, on the other hand, do not agree with such simple model. This differs from observations at lower temperature, between α - and ϵ -Fe, during both static loading at 300 K [20] and dynamic loading at the nanosecond timescales [22] where orientation relationships in agreement with a Burgers-type mechanism were observed. From the present result, it is unclear whether this complexity in transformation textures towards ϵ -Fe is related to variant selection, another transformation mechanism, or nucleation. This effect of starting crystallographic phase and temperature on transformation microstructures in ϵ -Fe will have to be investigated in the future.

Regarding the Earth core, it has been suggested that phase transformations in Fe towards a cubic phase could occur [45] and explain enigmatic observations of an innermost inner core [46]. Textures are important in inner core studies as they affect the shape of the measured seismic waves and induce an anisotropy in wave velocities that can be observed at a global scale [47]. To that regard, transformation textures in Fe should be included in core formation model [48]. Our results are very preliminary and cannot be integrated in such model yet but they pave the way towards a more materials science based understanding of this enigmatic portion of our planet.

5 Conclusions

An in situ study of stress and textures in polycrystalline bcc-, fcc-, and hcp-Fe has been carried out during plastic deformation up to 36 GPa and 1000 K in a DAC. The start of the phase transformation between α -Fe and γ -Fe is induced at 7 GPa and 973 K. Pressure is then increased up to 24 GPa at ~ 980 K, with a gradual transformation of α -Fe into γ -Fe and. At 14.5 GPa, α -Fe completely disappears and, above 14.5 GPa, we observe the gradual transformation of γ -Fe into ϵ -Fe. Temperature is then reduced to 900 K up to pressures of 33 GPa and to 300 K at 36 GPa, resulting in further transformation of γ -Fe into ϵ -Fe.

At 7 GPa and 973 K, α -Fe displays a texture with maxima at 001 and 011 with a marked minimum at 111, typical of compression textures in bcc metals. This texture is attributed to dominant slip on $\{110\}\bar{1}11$. Upon phase transformation γ -Fe develops a texture with a maximum near 011 and minima at 001 and 111. This texture is shown to be fully induced by the phase transformation, inherited from that of α -Fe, with very little effects of plastic deformation. Above 14.5 GPa, ϵ -Fe appears with a texture characterized by a broad maximum

291 30° from 0001 and a pronounced minimum near $10\bar{1}0$. This texture is the result of a phase
292 transformation from γ -Fe combined with plastic deformation with dominant basal
293 (0001) $\langle 11\bar{2}0 \rangle$ slip. Under differential stresses on the order of 0.2 to 0.6 GPa, it is shown that
294 the effect of plastic deformation must be accounted for in order to understand the texture
295 observed in ϵ -Fe, as the correspondence between transformation textures from γ -Fe or α -Fe
296 and experimental observations is poor.

297 The differential stress measured in ϵ -Fe is 0.6 ± 0.1 GPa at 24 GPa and 980 K, 1.3 ± 0.1 GPa
298 at 33 GPa and 900 K and 3.0 ± 0.4 GPa at 36 GPa and 300 K. These results, combined with
299 previous measurements of stress in ϵ -Fe indicate that, up to 1000 K and 55 GPa, the strength
300 of ϵ -Fe is ~ 1.9 GPa at 17 GPa and 300 K with a temperature and pressure dependence of -
301 0.003 GPa.K^{-1} and 0.074, respectively.

302 It is interesting to note that, with 10% axial strain, temperatures of ~ 900 K, and differential
303 stresses of 1.3 GPa, ϵ -Fe develops strong textures that are the result of plastic deformation.
304 This implies that, in the conditions of the Earth core, where stresses are probably low but
305 strains can be significant, ϵ -Fe can develop strong LPO that could be the source of the
306 observed seismic anisotropy [47].

307 The paper also demonstrates the capability of *in-situ* mapping of texture and stress over a
308 wide range of pressure and temperature. It could be applied to the *in-situ* study of orientation
309 relationships and variant selection during the wide range of phase transformations that can be
310 triggered when both pressure and temperature are varied, with numerous applications in
311 materials science, fundamental physics, and geophysics.

The development of new methods, such as in-situ three dimensional X-ray diffraction (3D-XRD) in the DAC [49] also offers great opportunities for understanding the physics of phase transitions under high pressure. The technique presented here can be used to study orientations in the polycrystal as a whole with good statistics. In parallel, 3D-XRD can be used to track precise orientation relationships between individual grains during the transformation. For instance, the transformation mechanism between γ - and ϵ -Fe is not well understood and a combination of both methods will be a powerful tool for deciphering the effect of mechanism, variant selection, and nucleation on the transition.

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Transformation	Model	Orientation relationships
fcc $\leftrightarrow$ bcc	Bain [15]	$\begin{Bmatrix} \{001\}_{\text{fcc}} \\ \langle 110 \rangle_{\text{fcc}} \end{Bmatrix} \parallel \begin{Bmatrix} \{001\}_{\text{bcc}} \\ \langle 100 \rangle_{\text{bcc}} \end{Bmatrix}$
fcc $\leftrightarrow$ bcc	Nishiyama-Wassermann [17]	$\begin{Bmatrix} \{111\}_{\text{fcc}} \\ \langle 01\bar{1} \rangle_{\text{fcc}} \end{Bmatrix} \parallel \begin{Bmatrix} \{110\}_{\text{bcc}} \\ \langle 001 \rangle_{\text{bcc}} \end{Bmatrix}$
fcc $\leftrightarrow$ bcc	Kurdjumov-Sachs [16]	$\begin{Bmatrix} \{111\}_{\text{fcc}} \\ \langle 110 \rangle_{\text{fcc}} \end{Bmatrix} \parallel \begin{Bmatrix} \{110\}_{\text{bcc}} \\ \langle 111 \rangle_{\text{bcc}} \end{Bmatrix}$
bcc $\leftrightarrow$ hcp	Burgers [23]	$\begin{Bmatrix} \{110\}_{\text{bcc}} \\ \langle \bar{1}\bar{1} \rangle_{\text{bcc}} \end{Bmatrix} \parallel \begin{Bmatrix} \{0001\}_{\text{hcp}} \\ \langle 11\bar{2}0 \rangle_{\text{hcp}} \end{Bmatrix}$
fcc $\leftrightarrow$ hcp	Shoji-Nishiyama	$\begin{Bmatrix} \{111\}_{\text{fcc}} \\ \langle 110 \rangle_{\text{fcc}} \end{Bmatrix} \parallel \begin{Bmatrix} \{0001\}_{\text{hcp}} \\ \langle 11\bar{2}0 \rangle_{\text{hcp}} \end{Bmatrix}$

399

400 **Table 1:** Orientation relationships for models of phase transformations between the bcc, fcc, and hcp structures. Please note that for the

401 Nishiyama-Wassermann model, the proposed orientation relationships are equivalent to $\{111\}\langle 2\bar{1}\bar{1} \rangle_{\text{fcc}} \parallel \{110\}\langle 1\bar{1}0 \rangle_{\text{bcc}}$.

402

Prismatic		Basal		Pyramidal <a>		Pyramidal <c+a>		Tensile twinning		Compressive twinning		
CRSS	Act. (%)	CRSS	Act. (%)	CRSS	Act. (%)	CRSS	Act. (%)	CRSS	Act. (%)	CRSS	Act. (%)	
Model 1	2	13.5	1	61.0	3	6.4	4	7.8	3	3.9	3	7.3
Model 2	0.7	0.3	0.1	99.1	*	*	1.2	0.2	0.8	0.4	*	*

Table 2: CRSSs and activities (Act) of the different deformation modes for model results shown in Fig. 5. Activities are shown at the end of the simulation (10% strain). CRSSs for model 1 are those of model A in Ref. [40]. CRSSs for model 2 are those of model 2 in Ref. [7] at 600 K after 0.3 accumulated strain in grain.

Figure 1: (Color online) Pressure-temperature path explored during the experiment. Grey-filled back circles indicate the observation of bcc iron (α -Fe), open blue squares of hcp iron (ϵ -Fe), and red diamonds of fcc iron (γ -Fe). γ -Fe coexists with ϵ -Fe past its stability limit until the end of the experiment. Solid lines are the iron phase diagram of [30]. Dashed line represents the α - γ phase boundary of [36]. Dot-dashed line shows the end of the co-existence domain of α - and ϵ -Fe, as determined at 300 K when no pressure medium is used [38].

Figure 2: “Unrolled” rXRD image (bottom) of γ -Fe and ϵ -Fe taken in-situ at 31 GPa and 900 K with the fit from Rietveld refinement (top). Diffraction lines from γ - and ϵ -Fe are labeled, and black arrows indicate the compression direction. Phase proportions are deduced from intensity ratios between both phases. The LPO and stress are deduced from the variations of diffraction intensity and peak position with orientation, respectively.

Figure 3: (Color online) Pressure, temperature, phase volume fractions, grain size, and differential stress in α - (grey-filled black circles), γ - (red filled diamonds) and ϵ -Fe (open blue squares) as a function of image number.

Figure 4: Inverse pole figures of the compression direction for α -, γ -, and ϵ -Fe representing the orientation of Fe polycrystals in the experiment. For each case, experimental pressure, temperatures, and phase proportions are shown on the figure. Numbers labeled with m and M indicate the minimum and maximum pole densities for each figure, respectively (in mrd). Equal area projection. Linear scale in mrd.

Figure 6: Inverse pole figures of the compression direction for α -, γ -, and ϵ -Fe representing the effects of phase transformation and plastic deformation on the orientations of Fe polycrystals. Bcc to fcc transformation is modeled according to the Nishiyama-Wassermann orientation relationships (N.W.). For the bcc to hcp transformation, we use a model based on the Burgers mechanisms. Finally, the fcc to hcp transformation is modeled according to the Shoji-Nishiyama orientation relationships (S.N.). The hcp texture obtained from the fcc phase is then used as a starting texture for VPSC simulations of plastic deformation up to 10% axial strain according to model 1 and 2 in Table 1. Equal area projection. Linear scale in mrd.

Table S1 : time, temperature, pressure, phase volume fractions, cell parameters, grain sizes, and differential stresses in bcc-, fcc-, and hcp-Fe as a function of image number.

Time (hours)	Image Number	T (K)	P (GPa)	Phase Fractions			Cell parameters (Å)					Grain size (Å)			Differential stress (GPa)		
				Bcc	Fcc	Hcp	a - Bcc	a - Fcc	a - Hcp	c - Hcp	c/a	Bcc	Fcc	Hcp	Bcc	Fcc	Hcp
0.0	003	295	0.1	100%	0%	0%	2.866					94			0.3		
1.4	013	295	0.2	100%	0%	0%	2.865					92			0.2		
2.9	022	295	1.5	100%	0%	0%	2.858					73			1.5		
3.7	024	295	3.4	100%	0%	0%	2.848					74			3.2		
4.8	028	295	4.6	100%	0%	0%	2.842					74			4.2		
5.6	031	295	4.8	100%	0%	0%	2.841					74			4.2		
15.7	035	373	12.8	42%	0%	58%	2.807		2.438	4.028	1.652	74		34	4.7		2.8
15.9	036	374	9.1	49%	0%	51%	2.823		2.454	3.978	1.621	74		61	4.8		3.6
16.3	037	374	5.9	54%	0%	46%	2.838		2.468	3.992	1.617	74		76	5.1		3.3
16.7	038	373	4.8	56%	0%	44%	2.843		2.473	4.096	1.656	109		119	4.5		2.9
17.7	040	474	8.6	53%	0%	47%	2.828		2.464	4.118	1.672	254		90	3.2		2.3
18.1	041	473	8.2	52%	0%	48%	2.830		2.468	4.117	1.668	252		91	3.2		2.1
18.4	042	474	6.9	53%	0%	47%	2.836		2.473	4.136	1.672	252		88	3.2		1.9
18.7	043	474	5.1	57%	0%	43%	2.845		2.477	4.151	1.676	252		77	3.1		1.8
19.4	045	570	4.1	81%	0%	19%	2.853		2.497	4.180	1.674	107		100	3.3		2.3
20.1	046	672	1.9	100%	0%	0%	2.869					122			0.8		
20.4	047	673	3.8	100%	0%	0%	2.858					139			0.9		
21.0	048	774	1.0	100%	0%	0%	2.878					378			0.0		
21.4	049	773	3.6	100%	0%	0%	2.863					431			0.3		
21.6	050	773	5.6	100%	0%	0%	2.852					386			0.5		
22.0	051	773	7.0	100%	0%	0%	2.845					343			0.5		
22.7	053	878	5.5	100%	0%	0%	2.856					581			0.1		
23.0	054	873	7.0	100%	0%	0%	2.848					454			0.1		
23.8	056	973	7.1	92%	8%	0%	2.851	3.619				506	167		0.0	0.2	
24.1	057	973	8.3	84%	16%	0%	2.845	3.606				470	281		0.4	0.2	
24.4	058	974	10.3	70%	30%	0%	2.835	3.580				571	168		0.2	0.1	
25.2	059	973	11.2	62%	38%	0%	2.831	3.566				473	176		0.2	0.1	
25.4	060	972	11.6	58%	42%	0%	2.829	3.558				469	138		0.2	0.0	
25.4	062	976	12.5	43%	57%	0%	2.825	3.546				424	168		0.2	0.1	
26.7	063	974	14.5	7%	89%	4%	2.818	3.529	2.481	4.030	1.624	424	368	201	0.5	0.2	
27.4	064	973	16.4	0%	92%	8%		3.519	2.476	4.012	1.620		238	242		0.4	0.3
27.8	065	975	18.0	0%	90%	10%		3.508	2.472	3.993	1.615		248	273		0.4	0.5
28.2	066	976	18.5	0%	89%	11%		3.504	2.470	3.990	1.616		228	293		0.3	0.4
28.7	067	976	19.9	0%	86%	14%		3.498	2.465	3.981	1.615		224	260		0.4	0.4
29.1	068	976	21.3	0%	85%	15%		3.491	2.460	3.970	1.614		190	289		0.4	0.6

29.4	069	976	22.9	0%	81%	19%	3.484	2.455	3.959	1.613	168	297	0.5
29.9	070	980	24.0	0%	73%	27%	3.476	2.451	3.953	1.613	148	279	0.6
30.3	071	953	25.2	0%	77%	23%	3.472	2.446	3.944	1.612	149	269	0.7
30.6	072	958	25.0	0%	63%	37%	3.470	2.447	3.946	1.613	129	258	0.5
31.0	073	958	25.6	0%	70%	30%	3.470	2.445	3.942	1.612	130	254	0.6
31.4	074	959	26.6	0%	68%	32%	3.465	2.442	3.936	1.612	124	259	0.8
31.8	075	938	26.4	0%	68%	32%	3.465	2.442	3.936	1.612	124	262	0.7
32.1	076	923	27.8	0%	72%	28%	3.465	2.438	3.924	1.610	109	427	1.4
32.4	077	903	28.2	0%	68%	32%	3.459	2.436	3.922	1.610	118	277	0.8
32.8	078	902	28.4	0%	61%	39%	3.455	2.435	3.921	1.610	113	267	0.9
33.1	079	901	29.4	0%	64%	36%	3.453	2.432	3.916	1.610	110	256	1.2
33.4	080	900	31.0	0%	59%	41%	3.444	2.427	3.908	1.610	110	262	1.3
33.8	081	899	33.2	0%	56%	44%	3.434	2.421	3.897	1.610	106	237	1.3
34.5	083	821	36.6	0%	56%	44%	3.419	2.410	3.876	1.608	92	203	1.5
35.2	085	707	35.4	0%	56%	44%	3.419	2.410	3.877	1.608	95	224	1.5
36.0	087	555	36.1	0%	61%	39%	3.413	2.406	3.865	1.607	90	243	1.7
36.4	088	422	36.0	0%	60%	40%	3.409	2.404	3.860	1.606	87	265	2.3
36.9	089	313	36.2	0%	66%	34%	3.410	2.402	3.855	1.605	86	395	3.6