

# Crystallographic texture formation in Fe-9wt%Si alloy during deformation and phase transition at high pressure

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## SUMMARY

The seismic anisotropy of the Earth's solid inner core has been the topic of much research. It could be explained by the crystallographic preferred orientation (CPO) developing during convection. The likely phase is hexagonal close-packed iron (hcp), alloyed with nickel and some lighter elements. Here we use high energy synchrotron X-rays to study CPO in Fe-9wt%Si, uniaxially compressed in a diamond anvil cell in radial geometry. The experiments reveal that strong preferred orientation forms in the low-pressure body-centred cubic (bcc) phase that appears to be softer than pure iron. CPO is attributed to dominant  $\{110\} \langle 111 \rangle$  slip. The onset of the bcc  $\rightarrow$  hcp transition occurs at a pressure of  $\approx 15$  GPa, and the alloy remains in a two phase bcc + hcp state up to 40 GPa. The hcp phase forms first with a distinct  $\{11\bar{2}0\}$  maximum perpendicular to compression. Modelling shows that this is a transformation texture, which can be described by Burgers orientation relationship with variant selection. Experimental results suggest that bcc grains oriented with  $\langle 100 \rangle$  parallel to compression transform into hcp first. The CPO of the hcp changes only slowly during further pressure and deviatoric stress increase at ambient temperature. After heating to 1600 K, a change in the hcp CPO is observed with alignment of (0001) planes perpendicular to compression that can be interpreted as dominant (0001) $\langle 11\bar{2}0 \rangle$  slip, combined with  $\{10\bar{1}2\} \langle \bar{1}011 \rangle$  mechanical twinning, which is similar to the deformation modes suggested previously for pure hcp iron at inner core conditions.

**Key words:** Composition and structure of the core; Phase transitions; Plasticity, diffusion and creep; Fe–Si; High-pressure behaviour.

## 1 INTRODUCTION

The solid inner core of the Earth has been of a long-standing interest in geophysics. Wiechert (1897) proposed a model for the Earth with an iron core and a stony shell to explain the average density. This was later supported by seismic observations (e.g. Oldham 1906; Gutenberg 1913) and observation of  $P$ -wave velocity reflections at a  $\sim 1400$  km radius. A jump in velocities led Lehmann (1936) to suggest an inner core with a higher density. Based on analysis of iron melting curves, Birch (1940) argued that the inner core should be solid, which was largely confirmed by refined  $P$ - and  $S$ -wave studies (e.g. Dziewonski & Gilbert 1971).

High pressure X-ray diffraction experiments revealed that with increasing pressure pure body-centred cubic (bcc) iron (A2 structure,  $\alpha$ -Fe) transforms to hexagonal close-packed (hcp) iron (A3 structure,  $\epsilon$ -Fe; e.g. Bancroft *et al.* 1956; Jamieson & Lawson 1962; Clendenen & Drickamer 1964; Takahashi & Bassett 1964). Since then, the phase diagram of iron was established over a wide range of pressures and temperatures, with both theory and experiments,

confirming stability of hcp iron at inner core conditions (e.g. Giles *et al.* 1971; Von Bagen & Boehler 1990; Brown 2001; Nishiyama *et al.* 2007; Tateno *et al.* 2010; Merkel *et al.* 2012, 2020; Anzellini *et al.* 2013; Ping *et al.* 2013; Godwal *et al.* 2015; Nishihara *et al.* 2018).

The presence of seismic anisotropy in the solid inner core of the Earth was first proposed based on velocity anomalies of  $P$ -waves that travel faster parallel to the Earth's rotation axis than in the equatorial plane (Morelli *et al.* 1986) and by splitting of inner core-sensitive oscillations (Woodhouse *et al.* 1986). It was suggested that this could be explained by alignment of anisotropic  $\epsilon$ -iron crystals due to deformation during solid-state convection (e.g. Jeanloz & Wenk 1988; Wenk *et al.* 2000a; Lincot *et al.* 2016), similar to convection in the mantle. Other possibilities include oriented growth during solidification (Bergman 1997; Deguen *et al.* 2011; Deguen 2012) and alignment in a magnetic field (Karato 1999; Buffett & Wenk 2001). The anisotropic seismic structure of the inner core was confirmed by many detailed studies and considerable local complexities were revealed (e.g. Deuss 2014; Souriau & Calvet 2015;

Tkalčić 2015; Romanowicz & Wenk 2017; Frost & Romanowicz 2019; Brett & Deuss 2020; Frost *et al.* 2021).

Consequently, much attention was attracted to plastic deformation of iron at increased temperatures and pressures. Near ambient conditions, bcc  $\alpha$ -Fe deforms largely by dislocation glide in the closest packed direction  $\{1\bar{1}0\}\langle 111\rangle$ , as revealed by deformation experiments, transmission electron microscopy and studies of texture development (e.g. Taylor & Elam 1926; Opinsky & Smoluchowski 1951; Watanabe 2006; Weinberger *et al.* 2013). At higher pressures, where hcp  $\epsilon$ -iron is stable, deformation experiments are usually conducted with diamond anvil cells (DAC) or multi-anvil apparatus (D-DIA), where anvils not only produce pressure, but also induce deviatoric stress that leads to formation of crystal preferred orientation (CPO) and can be used to deduce slip mechanisms (e.g. Mao *et al.* 2008; Miyagi *et al.* 2008; Merkel *et al.* 2012; Nishihara *et al.* 2018). Such experiments inferred basal (0001) and subordinate prismatic  $\{11\bar{2}0\}$  slip, combined with mechanical twinning as potential mechanisms for hcp Fe deformation (e.g. Wenk *et al.* 2000b; Merkel *et al.* 2004, 2013).

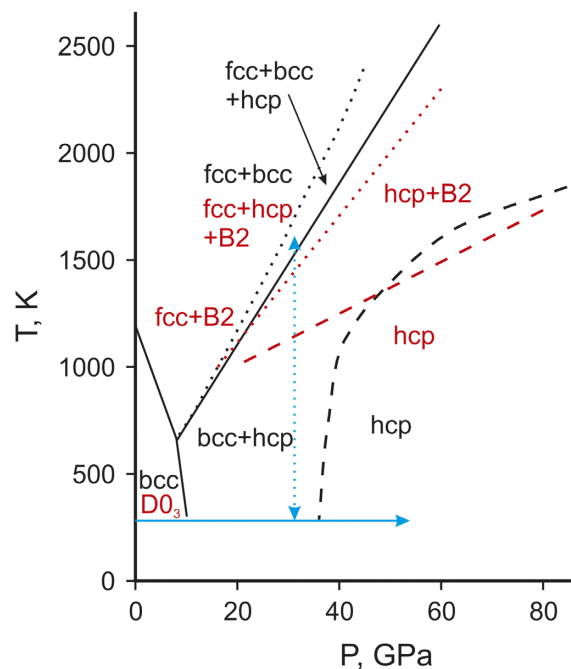
Birch (1952) was the first to recognize that the density of pure iron and iron–nickel alloy was too high and proposed the presence of  $\sim 10$  percent of lighter elements with carbon and silicon as candidates. Subsequently, different models explored the content of light elements, mainly oxygen, carbon, silicon and sulphur in the core (e.g. Alfè *et al.* 2002; Sata *et al.* 2010; Hirose *et al.* 2013; Morard *et al.* 2013; Badro *et al.* 2014; Ozawa *et al.* 2016). But  $P$ - and  $S$ -wave velocities of Fe–Si alloys remain too high compared with seismic observations and there are discrepancies between velocities and densities that still need to be resolved (e.g. Edmund *et al.* 2019; Nakajima *et al.* 2020).

In this context, Fe–Si systems with low Si content (less than about 10 weight percent  $\approx 18$  atomic percent) have been studied at high pressures (e.g. Lin *et al.* 2002, 2003; Hirao *et al.* 2004; Lin *et al.* 2009; Fischer *et al.* 2012; Fischer *et al.* 2013; Brennan *et al.* 2021) up to inner core conditions (Tateno *et al.* 2015). Depending on PT-conditions and Si content, Fe–Si alloys may crystallize in a variety of crystal structures (Cui & Jung 2017). A majority of DAC experiments observed a disordered bcc structure for low Si content Fe–Si alloys at room temperature and low pressures (e.g. Lin *et al.* 2002; Dubrovinsky *et al.* 2003; Lin *et al.* 2003; Hirao *et al.* 2004; Asanuma *et al.* 2008; Lin *et al.* 2009; Brennan *et al.* 2021). Ordered structures are also possible, for example a  $\text{DO}_3$  structure has been reported for a Fe-9wt%Si alloy at ambient conditions (Fischer *et al.* 2013, 2014). On pressure increase, similarly to pure iron, these cubic structures transform to the hcp phase (Fig. 1), though alloying with Si stabilizes the bcc structure of iron to higher pressures (Belonoshko *et al.* 2017; Edmund *et al.* 2019). It is argued that at inner core conditions Fe–Si alloys with relatively low Si content may consist of a single hcp phase (Kuwayama *et al.* 2009; Lin *et al.* 2009; Tateno *et al.* 2015; Komabayashi *et al.* 2019).

In this study, we use synchrotron X-ray diffraction in radial diamond anvil cell (rDAC) geometry to deform an iron–silicon alloy at high pressure and explore its deformation mechanisms, with implications for predicting elastic anisotropy during geodynamic movements in the inner core.

## 2 EXPERIMENTAL

The Fe-9wt%Si alloy (9 weight percent Si equals to 16.4 atomic percent) was selected for diamond-anvil cell (DAC) experiments in rDAC geometry. The powder sample (Goodfellow) was obtained



**Figure 1.** Phase diagram and PT paths of our experiments (blue). Solid phase boundaries are for the pure iron, black dashed and dotted lines and corresponding black labels are for Fe-7.9wt%Si [Lin *et al.* 2002], red dashed and dotted lines and corresponding red labels are for Fe-9wt%Si (Fischer *et al.* 2013). Blue arrows show PT paths of experiments with Fe-9wt%Si: Run #1 is the compression at ambient temperature (solid line), and Run #2 is also in compression, but with interim heating to 1600 K at constant pressure (dotted line).

from Kei Hirose and is the same material used in the Tateno *et al.* (2015) experiments. It was first tested with the SEM for compositional homogeneity.

DAC diffraction experiments in radial geometry were performed on end-station 2 of beamline 12.2.2 (Kunz *et al.* 2005) of the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory (LBNL) in 2016 and 2017. Fine powdered samples were loaded into a BX90 DAC (Kantor *et al.* 2012) with 300  $\mu\text{m}$  culet diamonds (Almax) using boron epoxy gaskets with inner diameter of 80  $\mu\text{m}$  inserted into supporting Kapton sheets. Loading was performed in a nitrogen atmosphere to suppress possible sample oxidation. A piece of platinum ( $\sim 15 \mu\text{m}$ ) was additionally loaded into the cell on top of sample grains (near one of the diamond culets) for Run #1 to be used as a pressure calibrant. Run #2 included laser heating of the sample, and we added nanocrystalline corundum powder with the sample at the contact with diamonds for Run #2 in order to enhance the thermal insulation between sample and diamonds and thus reduce temperature gradients in the sample. In Run #1 original sample thickness was 30  $\mu\text{m}$  that was reduced to 20  $\mu\text{m}$  over 3 hr. The sample thickness in radial diffraction was determined by performing absorption scans across the sample during alignment. In Run #2 the thickness was 50  $\mu\text{m}$ , part of it was corundum on bottom and top of sample. This did not change significantly during the 15 hr experiment.

Samples were aligned using absorption contrast scans at low X-ray energies (12 keV), while diffraction experiments were done at 25–30 keV (see Table 1 for details). The X-rays were focused to a spot-size of  $\sim 10$ –30  $\mu\text{m}$  using a Kirkpatrick Baez mirror set-up and the beam was aligned on the centre of the sample. Diffraction images were recorded with a marXperts MAR345 detector with

**Table 1.** Details of individual experimental runs.

| Run | Sample to detector distance (mm) | Energy (keV) | Beam size ( $\mu\text{m}^2$ )                       | Pressure standard                    | Additive                                   | Temperature (K)                   |
|-----|----------------------------------|--------------|-----------------------------------------------------|--------------------------------------|--------------------------------------------|-----------------------------------|
| #1  | 332.99                           | 25           | 30 × 30<br>(reduced to 10 × 10 at higher pressures) | Pt                                   | None                                       | 300                               |
| #2  | 330.43                           | 30           | 30 × 30                                             | Fe-9wt%Si (EOS based on Run #1 data) | Corundum for thermal insulation of diamond | 300; 1600 temporarily at 31.3 GPa |

345 mm diameter and 0.1 mm pixel size, located at a distance of ~330 mm. Exposure times varied from 60 to 150 s (when smaller beam cross-section was used at higher pressures, Table 1).

The load on a cell during the diffraction experiment was controlled using a gas membrane located at one end of the loading frame, similar to the setup described by Miyagi *et al.* (2008, their fig. 2). During Run #2 the sample was heated and then allowed to cool at a pressure of 31.3 GPa (Fig. 1) using the double-sided laser heating set-up at beamline 12.2.2, modified for *in situ* laser heating in radial diffraction (Kunz *et al.* 2018). The two laser spots were aligned on top of each other. Each laser heated hotspot has a full width at half-maximum (FWHM) of ~30  $\mu\text{m}$ . In order to ensure the whole sample to be exposed to high temperature, the sample was raster scanned across the two laser spots. Besides uniform exposure to high temperatures, this also ensures equilibration of pressure gradients possibly present in an unheated DAC without any pressure medium. As shown by Yen *et al.* (2020), during the process of laser heating a localized increase in pressure by several GPa can be observed within the hotspot with fairly steep pressure gradients across the hotspot. This pressure gradient is coupled to the presence of the hotspot and is released after temperature quench.

Diffraction images at selected pressures are displayed in Fig. 2.

Instrumental parameters (sample to detector distance, detector centre, detector tilt relative to incoming beam, X-ray energy and resolution function of the diffractometer) were determined by analysing the powder diffraction pattern of a standard  $\text{CeO}_2$  powder.

Complete analysis of all diffraction images was made following the procedures described by Lutterotti *et al.* (2014) and Wenk *et al.* (2014). Each image was segmented azimuthally over  $\eta = 5^\circ$  intervals and integrated into 72 conventional diffraction patterns. Patterns featuring bright diffraction spots from diamonds (1–3 on some images) were excluded from further analysis. All remaining patterns were simultaneously processed with the Rietveld analysis in MAUD software (Lutterotti *et al.* 1997). The refinement range was  $q \approx 2.6\text{--}5.8 \text{ \AA}^{-1}$  in the scattering vector magnitude scale ( $q = 2\pi/d$ , where  $d$  is the lattice spacing of diffraction planes). Phase volume fractions and unit cell parameters of all sample constituents, including platinum in Run #1 and corundum in Run #2, were refined. For each observed Fe-9wt%Si phase, atomic content of silicon was fixed at 16.4 atomic percent.

Diffacted intensity variations along Debye rings (Fig. 2) are indicative of preferred orientation of crystallites. The discrete E-WIMV algorithm (Matthies 2002; Lutterotti *et al.* 2014) was used to calculate orientation distribution functions (ODFs) of all phases. Sets of diffraction patterns obtained at pressures above ~4 GPa were found to be in agreement with fibre symmetry of the ODF

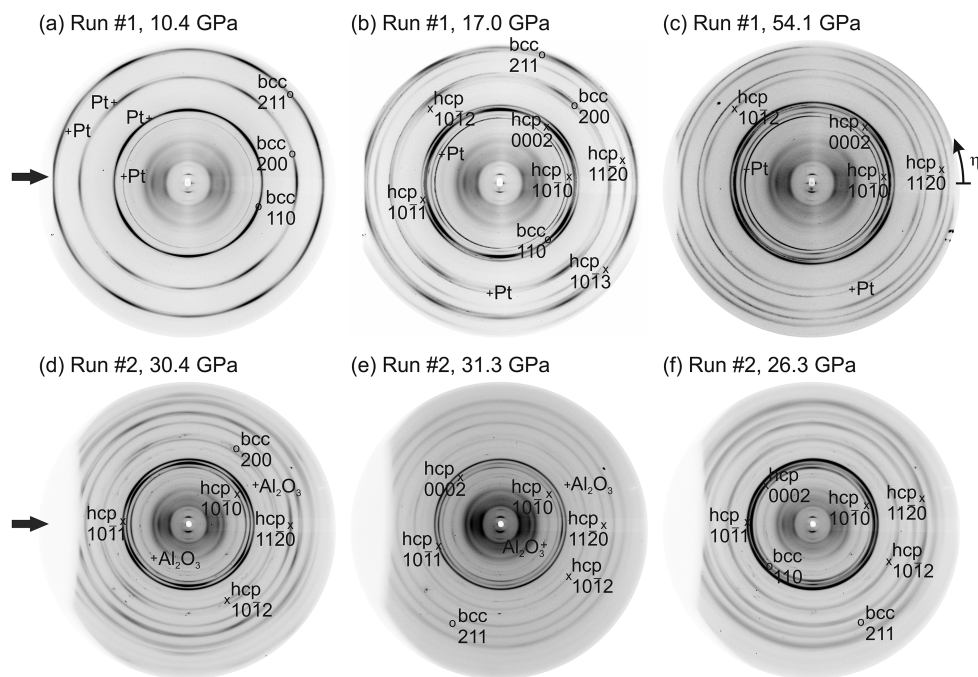
around the compression axis corresponding to the DAC deformation geometry, and a fibre symmetry and  $5^\circ$  cell size in orientation space were used in the E-WIMV algorithm. Recalculated ODFs were exported from MAUD into the BEARTEX program (Wenk *et al.* 1998) for smoothing with a Gaussian function with full width at half-maximum of  $7.5^\circ$  and inverse pole figures (IPFs) of the compression direction were recalculated from the smoothed ODFs. Inverse pole figures (displayed in Figs 5, 9 and 10) illustrate the orientation of a sample direction (compression axis) relative to crystal coordinates. Through this paper, only IPFs of the compression direction are discussed, thus the term ‘compression direction’ is omitted in most of the following text.

According to the accepted refinement procedure (Lutterotti *et al.* 2014), macroscopic stresses in the rDAC are separated into a hydrostatic and a deviatoric part. The hydrostatic part (pressure) only influences unit cell parameters. The deviatoric part of the macroscopic stress tensor is diagonal with a zero trace, where  $\sigma_{11} = \sigma_{22} = -\sigma_{33}/2$  (assuming that  $\sigma_{33}$  is the stress along the compression axis of the DAC), and all other components are equal to zero. The differential stress value  $t = \sigma_{33} - \sigma_{11}$  is related to maximum shear or von Mises yield criterion (Ruoff 1975; Singh 1993), and in the following text the absolute  $t$  value is used (in GPa). We used the ‘Moment Pole Stress’ model in MAUD and for the bcc and hcp Fe-9wt%Si alloy pressure-dependent elastic constants of the bcc and hcp Fe-6.7wt%Si alloy (Tsuchiya & Fujibuchi 2009) to calculate stress.

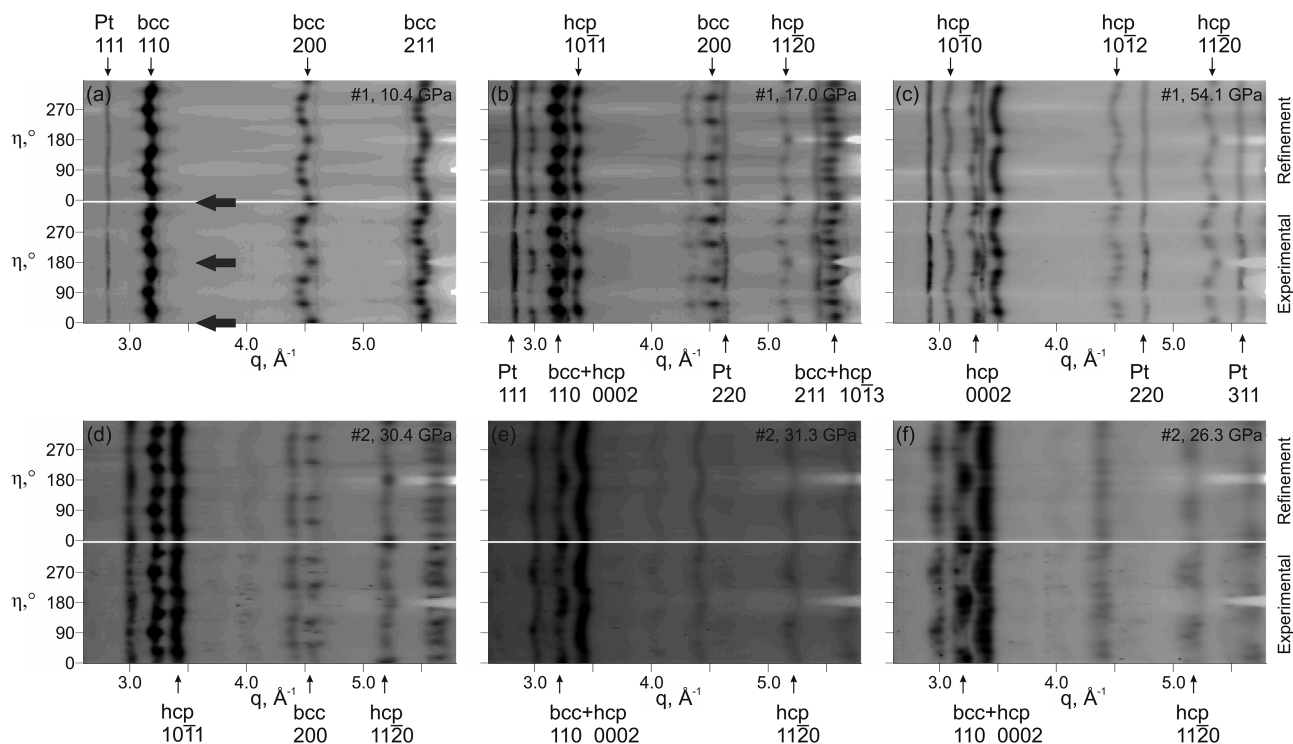
Deviatoric stresses change the shape of Debye rings from circles to ellipses, meaning that positions of diffraction peaks shift from pattern to pattern. Consequently, we recalculated stress values from observed peak shifts using the ‘BulkPathGEO’ model (Matthies *et al.* 2001) (labeled as ‘Moment Pole Stress’ in MAUD) that considers CPO and single crystal elastic constants of the material. Pressure dependencies of elastic coefficients of platinum (Menéndez-Proupin & Singh 2007) were used for Run #1 data analysis and of corundum (Duan *et al.* 1999) in Run #2.

Pressure in the DAC during Run #1 was calculated using equation of state of platinum (Matsui *et al.* 2009). Due to overlap of platinum diffraction peaks with peaks of Fe-9wt%Si alloy phases, in Run #2 pressure calibration was made using the equation of state of Fe-9wt%Si obtained by fitting Run #1 data with the Birch-Murnaghan equation of state (Birch 1947).

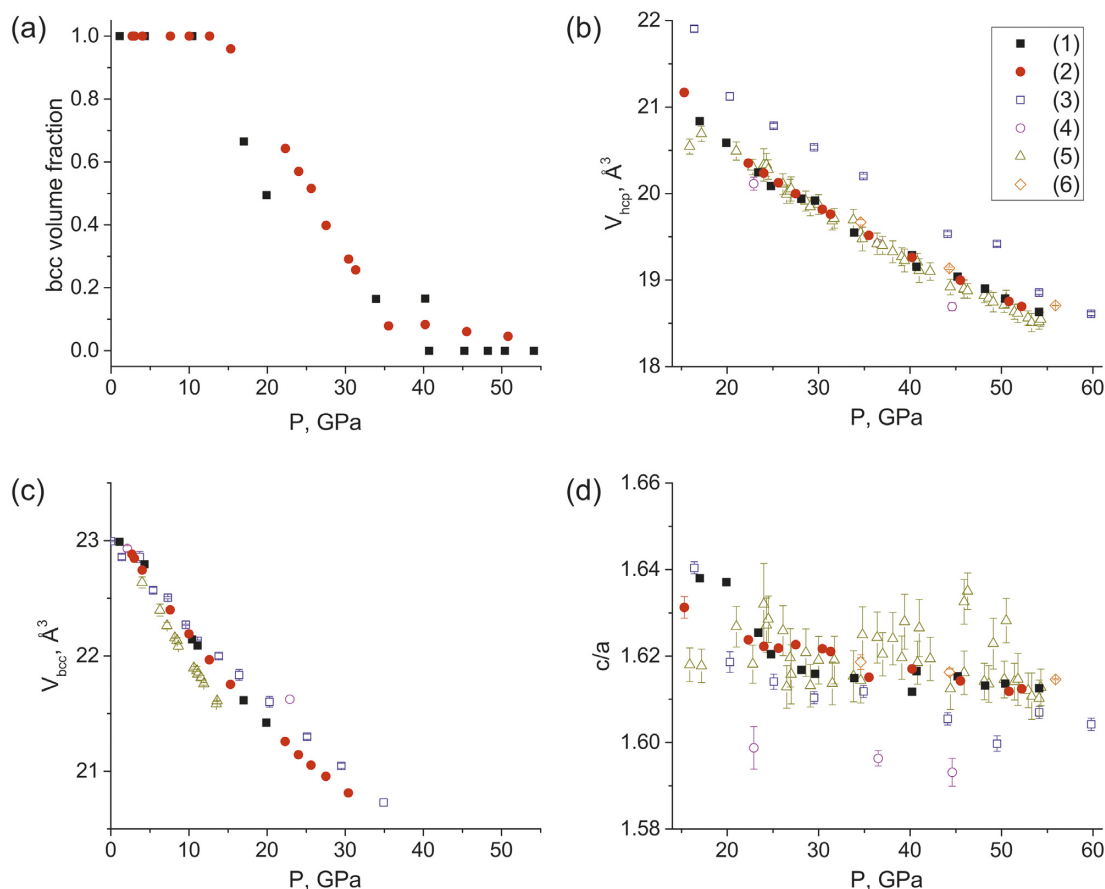
Several analysed ‘unrolled’ diffraction images as function of azimuth, corresponding to Fig. 2, are shown in Fig. 3. A good agreement of experimental and refined diffraction intensities and peak position variations is observed, indicating reliability of the fit.



**Figure 2.** Characteristic diffraction images of Runs #1 and #2 at different stages of the experiment: (a) single phase bcc Fe-9wt%Si; (b) two-phase bcc + hcp mixture; (c) single phase hcp; (d) two-phase bcc + hcp mixture prior to heating during Run #2; (e) at 1600 K temperature; (f) during unloading. Intensive diffraction rings of bcc (o) and hcp (x) phases of Fe-9wt%Si are indexed. Rings of platinum (Run #1) and corundum (Run #2) are also shown (+). Note that some diffraction rings, for example hcp 100 and bcc 110 largely overlap. Block arrows show the compression direction.



**Figure 3.** Unrolled diffraction images, corresponding to Fig. 2, with azimuth  $\eta$  as a function of scattering vector magnitude  $q$ . Lower half of every figure is experimental data, upper half is refinement result. Some of the intensive diffraction peaks are indexed. Note, that some of diffraction patterns, featuring intensive peaks from scattering on diamonds, were disabled for the refinement and are not plotted here. Block arrows show compression direction.



**Figure 4.** Pressure dependencies of Fe-Si alloy parameters. (a) bcc phase volume fraction; (b) hcp phase unit cell volume; (c) bcc phase unit cell volume; (d) hcp phase  $c/a$  ratio. In (b), (c) and (d), data for Fe-Si alloys with close composition are also shown for comparison. (1) Fe-9wt%Si, this work, Run#1; (2) Fe-9wt%Si, this work, Run#2; (3) Fe-8.7wt%Si (Hirao *et al.* 2004), their Run 1; (4) Fe-8.7wt%Si (Hirao *et al.* 2004), their Run 2; (5) Fe-7.5wt%Si (Lin *et al.* 2003), during the pressure increase; (6) Fe-9wt%Si (Tateno *et al.* 2015), their #4 during the pressure increase.

After the Run #2 experiment, the deformed sample in the gasket was recovered from the DAC and studied with a Zeiss EVO SEM at 20 kV and 11k magnification. The chemical composition was evaluated with an EDAX EDS system.

### 3 RESULTS

#### 3.1 Observed Fe-9wt%Si phases and unit cell parameters

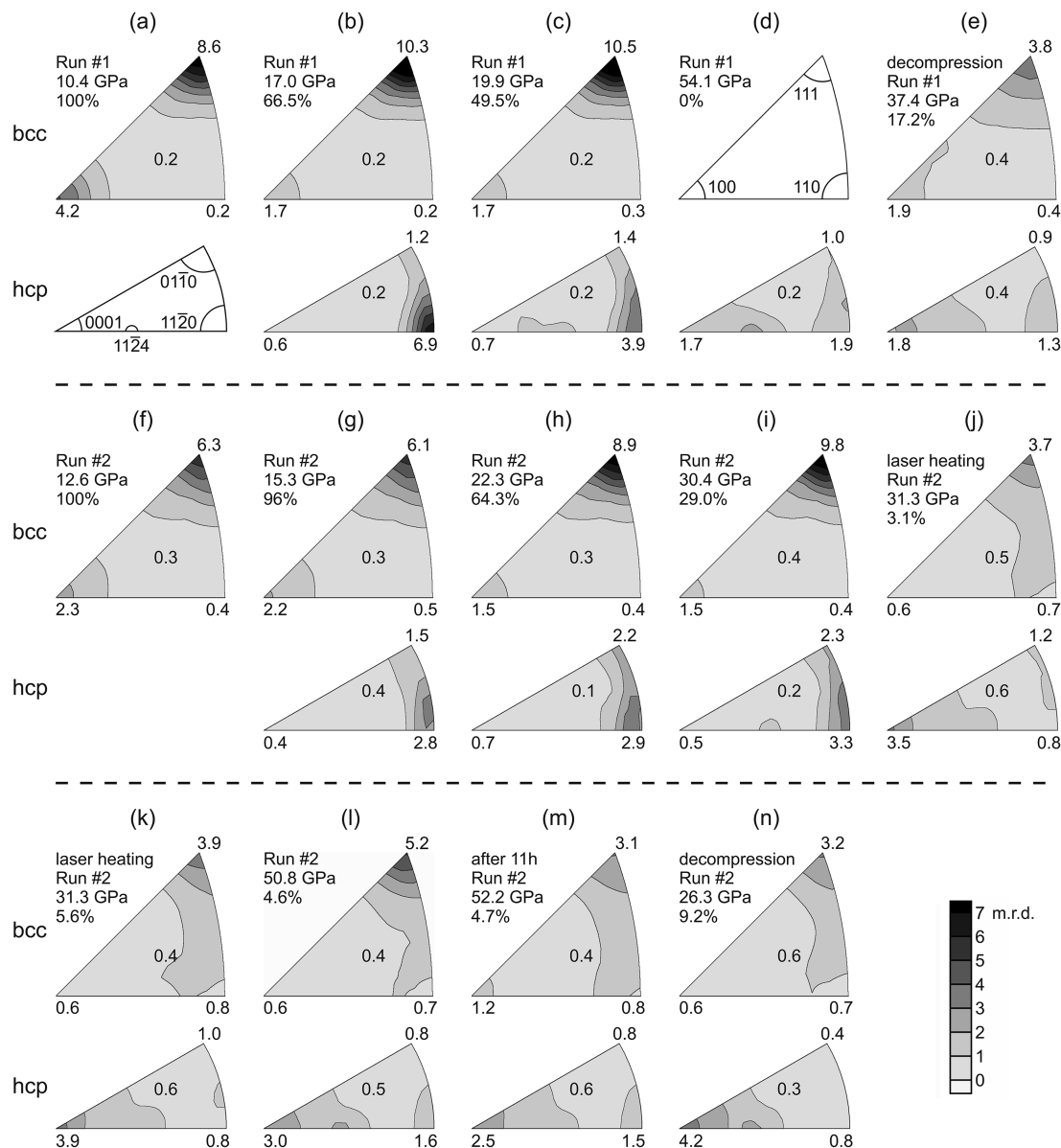
Our data indicate that Fe-9wt%Si samples are in a single bcc phase state at ambient conditions and pressures below  $\sim 15$  GPa. There was one exception found during Run #2 at the starting pressure of 2.7 GPa. Additional weak diffraction peaks were observed that could be attributed either to the B2 phase as superstructure reflections 111 and 210, or to the  $D0_3$  phase as peaks 222 and 420 (Fig. S1). Traces of the 311 diffraction peak of  $D0_3$  may be poorly manifested due to overlap with corundum reflection, and the 331 peak is not visible in diffraction patterns, but it may be due to its low intensity. These peaks disappeared during further pressure increase.

With pressure increase, the bcc phase starts transforming into hcp. The earliest onset of hcp diffraction peaks was observed at a pressure of 15.3 GPa (Run #2), where the refinement converges to 4 volume percent of hcp Fe-9wt%Si. At higher pressures, the bcc phase content gradually decreases, and at pressures over  $\approx 20$  GPa, hcp becomes the dominant phase. During Run #1, the sample was

found to have completely transformed to hcp phase at a pressure of 40.7 GPa. During Run #2, minor ( $\sim 5$  vol percent) amounts of the bcc phase persisted up to the maximum pressure of 52.4 GPa (Fig. 4a). Part of this may be due to pressure gradients in the DAC, because of the averaging over whole circular sample in rDAC experiment with a diffraction signal that includes sample both at high and low pressure. In Run #1, this effect was suppressed due to reduced beam cross-section at higher pressures (Table 1), which left out of the beam most of the sample exterior.

The unit cell volumes of Fe-9wt%Si phases, and  $c/a$  ratio for the hcp Fe-9wt%Si obtained from Run#1 data with error bars are plotted in Fig. 4 as functions of pressure and compared with data from the literature. Unit cell parameters of the phases are given in Table 2.

Equations of state (EOS) of the bcc and hcp Fe-9wt%Si phases were obtained based on Run #1 data. Only the pressure range with the respective phase volume fractions over  $\sim 0.3$  was used, where the diffraction peaks of the phase have sufficient intensity, and unit cell parameters could be most reliably determined. Equations of state of Fe-9wt%Si compressed in rDAC were obtained by fitting unit cell volume dependencies on pressure with the Birch–Murnaghan equation (Birch 1947), since the influence of non-hydrostatic stresses on the equation of state of Fe-9wt%Si at ambient temperature is insignificant (Fischer *et al.* 2014). Pressure ranges of 1.1–19.9 GPa and 17.0–54.1 GPa were used for bcc phase and hcp phase, respectively. Resulting EOS parameters



**Figure 5.** Selected inverse pole figures of bcc and hcp Fe-9wt%Si. Equal area projections, linear scale. Corresponding crystal coordinate systems for bcc and hcp phases are shown in the first two rows. Run number, pressure and bcc phase volume content are also provided in addition to peak intensities in main IPF directions; number inside the IPF indicates minimum pole density (m.r.d.; rounded to one decimal digit).

for bcc Fe-9wt%Si are  $K_0 = 205(8)$  GPa,  $V_0 = 23.18(4)$  Å<sup>3</sup> (or  $6.98(1)$  cm<sup>3</sup> mol<sup>-1</sup> atom<sup>-1</sup>), and the  $K_0'$  parameter was fixed to 5.26, the value, obtained by Lin *et al.* (2003) for the bcc Fe-7.9wt%Si alloy, to ensure the refinement convergence in a relatively narrow pressure interval. For hcp Fe-9wt%Si we set  $K_0 = 165(7)$  GPa,  $V_0 = 22.53(10)$  Å<sup>3</sup> (or  $6.78(3)$  cm<sup>3</sup> mol<sup>-1</sup> atom<sup>-1</sup>), and the  $K_0'$  parameter was fixed to 5.7 (Lin *et al.* 2003).

Obtained EOS were used to determine the pressure in the DAC during Run #2, the bcc EOS at pressures below 20 GPa, and the hcp EOS at pressures over 20 GPa. Therefore, our Run #2 unit cell volume dependencies on pressure exactly repeat Run #1 data.

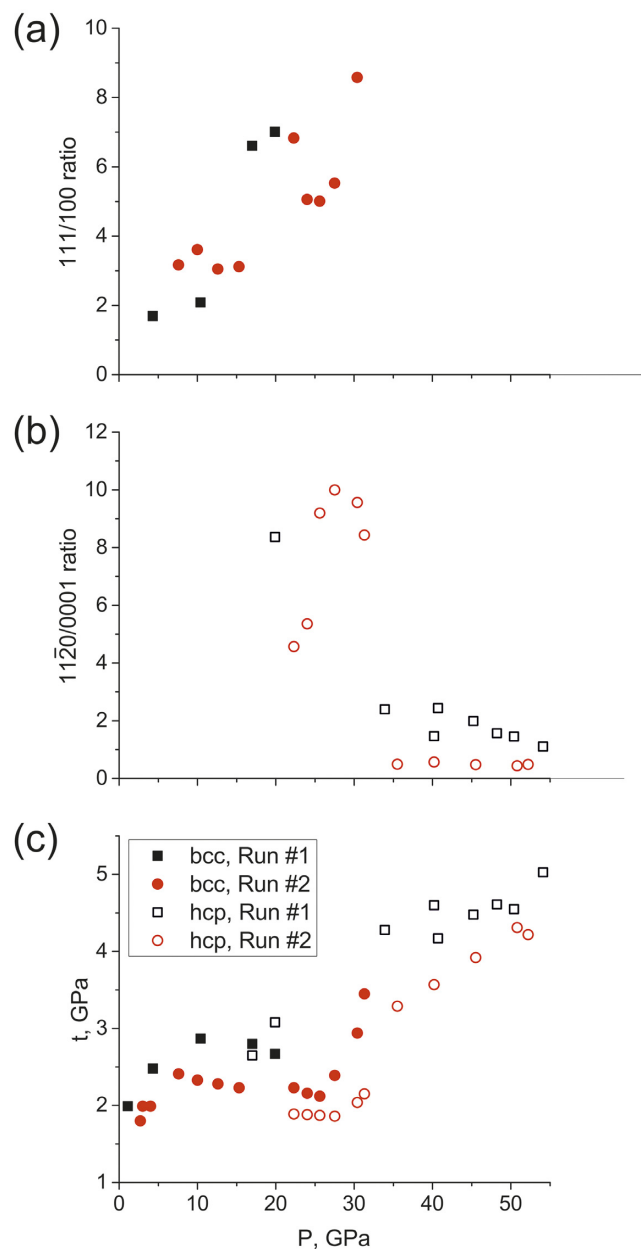
Only diffraction peaks of the bcc and hcp Fe-9wt%Si phases were observed. Even during heating at a pressure of 31.3 GPa in Run #2 and temperature of 1600 K, there was no evidence for an fcc phase.

During the sample decompression, only bcc and hcp Fe-9wt%Si phases were observed. Corresponding diffraction rings during the

decompression are more broad and diffuse (Fig. 2f), indicating large stress gradients in the DAC, and making reliable determination of pressure values, deviatoric stresses and unit cell parameters difficult. Thus, decompression results were not used for Table 2 or plots in Fig. 4.

### 3.2 Crystal preferred orientations and deviatoric stresses

Information on CPOs of bcc and hcp Fe-9wt%Si is given as inverse pole figures (IPF) of the compression direction (Fig. 5). Texture maxima and minima are highlighted in each IPF. The initial texture of the bcc phase before compression in the DAC (not shown in Fig. 5) is irregular and attributed to a sample composed of a limited number of grains, with a few larger ones dominating the CPO pattern. Consequently, texture is characterized by the absence of a



**Figure 6.** Pressure dependencies of Fe-9wt%Si phases parameters: (a) ratio of IPF pole density values at (111) and (100) after the random texture component subtraction for the bcc phase; (b) ratio of IPF pole density values at (1120) and (0001) after the random texture component subtraction for the hcp phase; (c) absolute  $t$  values for bcc and hcp phases.

random component (minimum ODF value is zero) and a relatively high ODF maximum.

#### Run #1 analysis

During Run #1, already at 4.3 GPa a symmetric preferred orientation pattern of bcc Fe-9wt%Si develops with two distinct maxima in the IPF (Fig. 5a), indicating significant plastic deformation with a corresponding differential stress value  $t = 2.6$  GPa (Fig. 6c, Table 2). The most intense texture component is at (111) with a value of 5.7 m.r.d. (multiple of a random distribution) and a secondary maximum is at (100) with 3.5 m.r.d. The peak intensity ratio is 1.69

(Fig. 6d), which is calculated after subtracting the contribution of a random CPO component (i.e. the minimum ODF value).

CPO sharpness of the bcc phase increases with subsequent pressure increase up to a (111) maximum of 8.6 m.r.d. at 10.4 GPa, and 10.5 m.r.d. at 19.9 GPa (Figs 5a and c). Correspondingly the minimum value decreases. Differential stress values for the bcc phase remain on a level of  $\sim 2$ –3 GPa (Fig. 6c). The beginning of the phase transformat

ion leads to a redistribution of strength of (111) and (100) texture components in IPFs (Figs 5b and c). Prior to the transformation, the peak intensity of the (111) component is only 1.5–2 times higher than the peak intensity of the (100) component. After the appearance of the hcp phase, this ratio increases to  $\approx 5$ –7, and the (111) component dominates the bcc CPO (Fig. 6a).

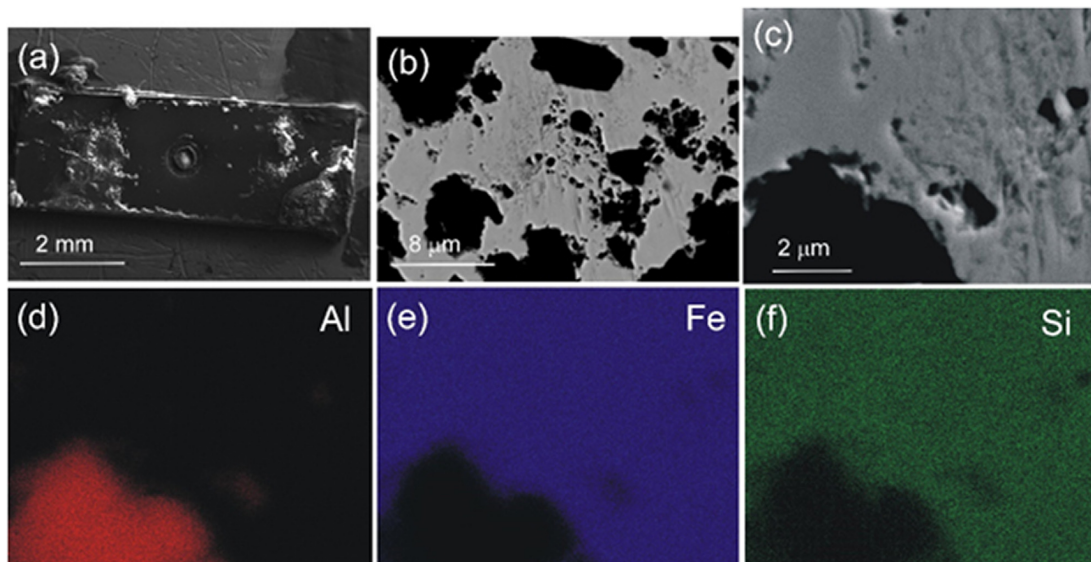
The hcp phase at 17 GPa immediately has a distinct maximum in IPF at (1120) (Fig. 5b). The maximum is strongest initially (6.9 m.r.d., 33.5 vol per cent of the hcp phase), but becomes more diffuse as the transformation progresses (3.8 m.r.d. at 23.4 GPa with 63.7 vol per cent hcp), and a secondary maximum develops in IPF at  $\approx 36^\circ$  to (0001), close to (1124) (Fig. 5c).

At higher pressures, when the sample is in a single hcp phase state, the CPO remains practically stable up to the maximum pressure of 54.1 GPa. IPF values at (1120) gradually decrease to 1.9 m.r.d. at 54.1 GPa, while values near (0001) slowly increase up to 1.70 m.r.d. (Fig. 5d). The ratio of these IPF components (Fig. 6b) remain over 1, indicating only a very slow development of an hcp deformation texture, despite an observed increase of differential stress to  $\approx 4$ –5 GPa range (Fig. 6c). That is higher than values reported for pure hcp iron (Merkel *et al.* 2005; Nishiyama *et al.* 2007; 2013) and some Fe–Ni–Si alloys (Brennan *et al.* 2021).

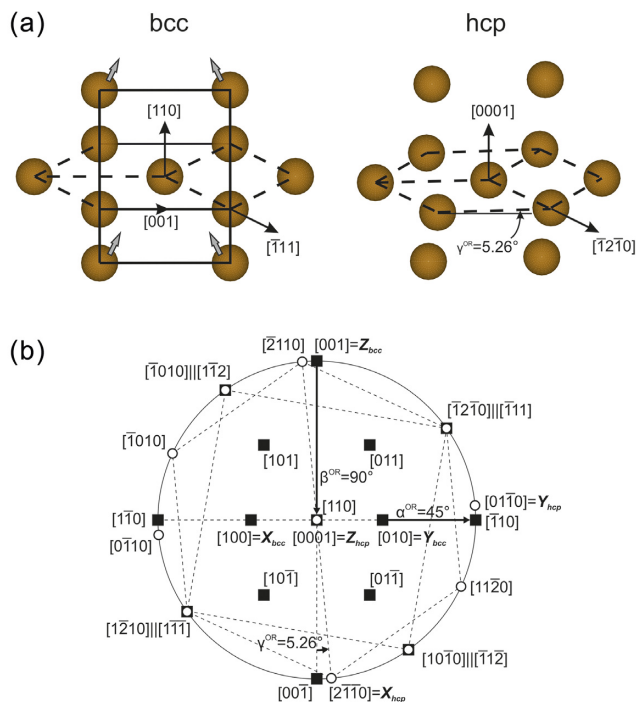
During decompression, bcc Fe-9wt%Si reappears at a pressure of 40.2 GPa. The CPO is weak, but with a distinct (111) component in IPF (Fig. 5e). It should be noted that the unit cell parameter of the bcc phase at this pressure is already close to its ambient conditions and  $t \approx 0.32$  GPa, indicating that the bcc phase is depressurized due to stress gradients in DAC. During further decompression the bcc texture becomes sharper and the (111) maximum increases to 3.8 m.r.d., and a weak (100) IPF component appears. The hcp texture during the decompression retains the same persisting (1120) component in IPFs. The auxiliary maximum close to (1124) broadens, and a component close to (0001) forms (Fig. 5e).

#### Run #2 analysis

During Run #2, a small pressure of 2.7 GPa was already introduced during the DAC assembly. Formation of the fibre CPO of the bcc phase is observed at 7.6 GPa ( $t = 2.41$  GPa) with the preferred orientation pattern very similar to Run #1 (Fig. 5f): there is one strong (111) maximum and a weaker (100) component in the compression direction. Overall pole densities of these components are lower than in Run #1. Similar to Run #1, the (100) component of the bcc texture becomes weaker with the formation of the hcp phase at 15.3 GPa (Figs 5g and h) and the ratio of pole densities at (111) and at (100) increases to over 5 (Fig. 6a). The hcp phase appears with the preferred orientation forming a broad IPF maximum close to (1120) (Fig. 5g). As in Run #1, in Run #2 the hcp texture remains rather stable with the increase of the hcp volume fraction and the maximum at (1120) broadens further, contributing some orientation intensity to (1010) and a weak maximum close to (1124) appears



**Figure 7.** SEM images of the recovered Run #2 sample. (a) Low magnification image of the Fe-9wt%Si sample in gasket on foil. (b) Backscatter electron (BE) image of the Fe-9wt%Si sample (bright) with corundum particles (dark). (c) BE image of area selected for EDS scans. (d–f) EDS scans of Al, Fe and Si documenting a uniform chemical composition of the Fe-9wt%Si alloy.



**Figure 8.** Schematic layout of the Burgers OR: atomic rearrangement (solid lines show the edges of the bcc unit cell, grey block arrows show shuffling direction of alternating (110) bcc planes) (a); stereographic projection of different lattice directions in the bcc (■) and hcp (○) phases arranged according to the Burgers OR (b). All three-index indices are the indices of the bcc phase directions, four-index indices are the Bravais indices of the hcp phase directions.

(cf. Figs 5c and i). A notable difference is that in Run #2 differential stress values in hcp phase are  $\approx 1$  GPa lower than in Run #1 (Fig. 6c). It is likely related to the presence of corundum during Run#2, resulting in redistribution of mechanical stresses.

As will be discussed later, the (11 $\bar{2}$ 0) hcp texture is attributed to the phase transition. In an attempt to erase the transformation texture of the hcp phase, the sample was heated to 1600 K during Run #2 at 31.3 GPa, where the bcc content became relatively low ( $\sim 20$  vol per cent). Diffraction images recorded during the heating document that the bcc texture does not change qualitatively under the increased temperature, but the hcp phase develops a CPO with (0001) IPF maximum over 3.5 m.r.d., while the (11 $\bar{2}$ 0) IPF component becomes depleted ( $\approx 0.8$  m.r.d., Figs 5j and k). Consequently, the ratio of these components drops well below 1 (Fig. 6b). During further pressure increase at room temperature, the formed hcp CPO remains weak and maintains the most intensive component at (0001) (Fig. 5l) and does not change significantly after an 11 hr sample exposure at a pressure over 50 GPa (Fig. 5m).

During decompression in Run #2, the symmetries of CPOs do not change significantly. Similar to Run#1, the main IPF components are at (111) for bcc and (0001) for hcp (Fig. 5n).

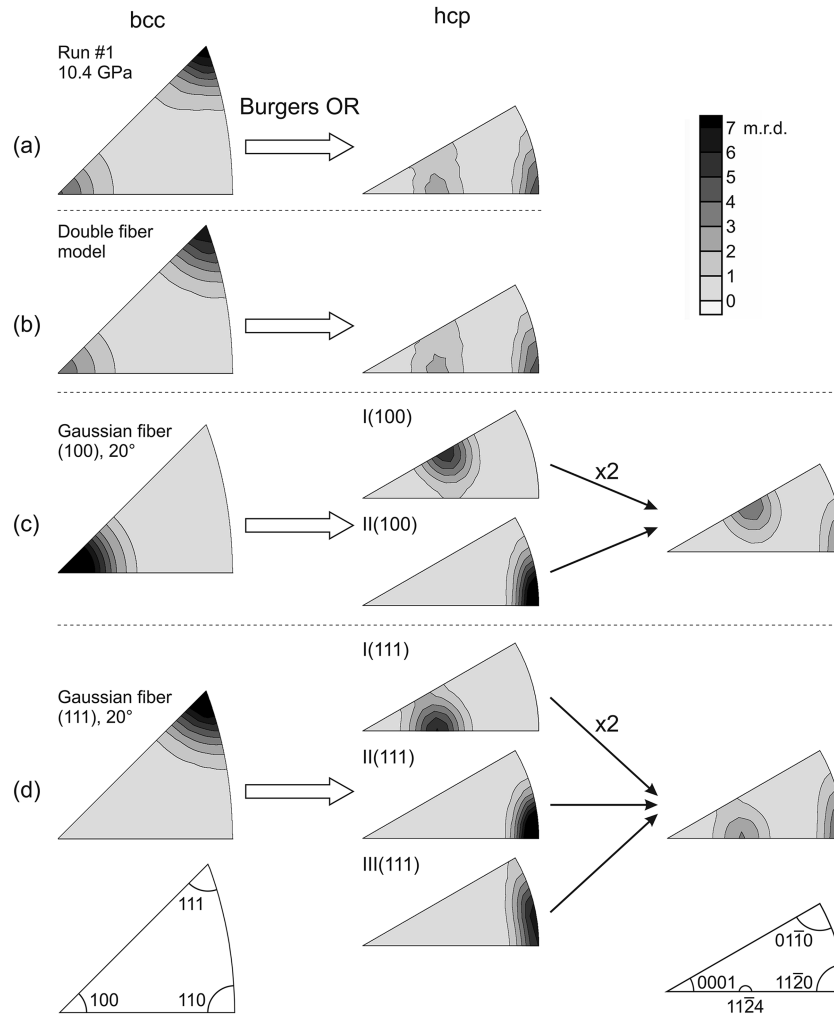
### 3.3 Chemical composition of the recovered sample

Backscatter SEM images (BE) of the sample recovered from Run #2 diffraction experiments show no evidence for phase separation (Figs 7b and c), nor do energy-dispersive spectroscopy (EDS) maps of elements (Figs 7d–f). The sample appears as a chemically homogeneous Fe–Si alloy with no evidence for different Fe–Si compositions. The Al signal (Fig. 7d) is from a grain of corundum.

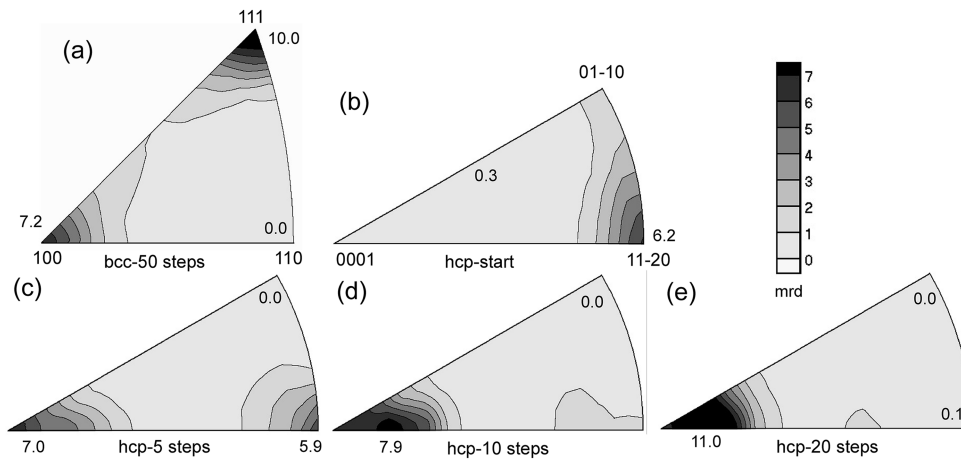
## 4 DISCUSSION

### 4.1 Composition, crystal structures and equations of state of Fe-9wt%Si alloy

The dissociation of Fe–Si alloys into Si-rich and Si-poor phases could happen when an Fe–Si alloy is simultaneously subjected to high pressures and temperatures, for example 45 GPa at 1650 K for Fe-9.9wt%Si (Kuwayama *et al.* 2009) or 93 GPa at 2100–2400 K for



**Figure 9.** Transformation texture models shown in IPFs. (a) Transformation of an experimental (Run#1, 10.4 GPa) bcc texture by applying Burgers OR; (b) Transformation of the same bcc texture, represented by a model with two 20° Gaussian fibres and a random component, by applying Burgers OR. (c) Transformation of the bcc 20° Gaussian fibre (100) by applying Burgers OR; two different variants of IPFs [I(100) and II(100)] and hcp texture without variant selection are shown. (d) Transformation of the bcc 20° Gaussian fibre (111) by applying Burgers OR; three different variants of IPFs [I(111), II(111), and III(111)] and hcp texture without variant selection are shown. Equal area projections, linear scale.



**Figure 10.** VPSC model of texture evolution during axial compression. Inverse pole figures, equal area projection. (a) bcc after 50 percent strain. (b) Starting texture used for hcp VPSC models. (c) hcp after 5 percent strain; (d) 10 percent strain and (e) 20 percent strain.

**Table 2.** Selected refined bcc volume fraction value, unit cell parameters, differential stress values  $t$ , of Fe-9wt%Si phases (only for corresponding phase volume fraction over  $\sim 0.3$ ). Total time the sample was in DAC is given in the first column (starting from first diffraction image measurements of the Run). Standard errors of the last digit are in parentheses.

| Time (hr) | $P$ (GPa) | bcc volume fraction | $a_{\text{bcc}}$ (Å) | $t_{\text{bcc}}$ (GPa) | $a_{\text{hcp}}$ (Å) | $c_{\text{hcp}}$ (Å) | $t_{\text{hcp}}$ (GPa) |
|-----------|-----------|---------------------|----------------------|------------------------|----------------------|----------------------|------------------------|
| Run#1     |           |                     |                      |                        |                      |                      |                        |
| 0.0       | 1.1       | 1.0                 | 2.8434(1)            | 1.99(2)                |                      |                      |                        |
| 0.1       | 4.3       | 1.0                 | 2.8354(1)            | 2.48(2)                |                      |                      |                        |
| 0.4       | 10.4      | 1.0                 | 2.8081(1)            | 2.87(2)                |                      |                      |                        |
| 0.6       | 17.0      | 0.665(1)            | 2.7856(1)            | 2.80(5)                | 2.4490(2)            | 4.0114(5)            | 2.65(3)                |
| 0.9       | 19.9      | 0.495(3)            | 2.7773(1)            | 2.67(2)                | 2.4397(1)            | 3.9940(4)            | 3.08(3)                |
| 2.5       | 33.9      | 0.165(12)           |                      |                        | 2.4089(1)            | 3.8901(2)            | 4.28(2)                |
| 2.6       | 40.2      | 0.166(24)           |                      |                        | 2.3997(1)            | 3.8677(4)            | 4.60(3)                |
| 2.8       | 40.7      | 0                   |                      |                        | 2.3918(1)            | 3.8664(2)            | 4.17(2)                |
| 2.9       | 45.2      | 0                   |                      |                        | 2.3875(1)            | 3.8566(2)            | 4.48(2)                |
| 3.0       | 48.2      | 0                   |                      |                        | 2.3828(1)            | 3.8439(2)            | 4.61(2)                |
| 3.2       | 50.4      | 0                   |                      |                        | 2.3778(1)            | 3.8369(2)            | 4.55(2)                |
| 3.3       | 54.1      | 0                   |                      |                        | 2.3717(1)            | 3.8245(3)            | 5.03(3)                |
| Run#2     |           |                     |                      |                        |                      |                      |                        |
| 0.0       | 2.7       | 1.0                 | 2.8390(1)            | 1.80(1)                |                      |                      |                        |
| 0.2       | 3.0       | 1.0                 | 2.8375(1)            | 1.99(1)                |                      |                      |                        |
| 0.3       | 4.0       | 1.0                 | 2.8333(1)            | 1.99(1)                |                      |                      |                        |
| 0.4       | 7.6       | 1.0                 | 2.8189(1)            | 2.41(1)                |                      |                      |                        |
| 0.6       | 10.0      | 1.0                 | 2.8101(1)            | 2.33(1)                |                      |                      |                        |
| 0.7       | 12.6      | 1.0                 | 2.8006(1)            | 2.28(1)                |                      |                      |                        |
| 0.8       | 15.3      | 0.960(1)            | 2.8390(1)            | 2.23(1)                |                      |                      |                        |
| 1.1       | 22.3      | 0.643(1)            | 2.8375(1)            | 2.23(2)                | 2.4370(1)            | 3.9572(6)            | 1.89(3)                |
| 1.2       | 24.0      | 0.57(1)             | 2.8333(1)            | 2.16(2)                | 2.4331(1)            | 3.9471(5)            | 1.88(3)                |
| 1.3       | 25.6      | 0.516(1)            | 2.8189(1)            | 2.12(2)                | 2.4289(1)            | 3.9393(5)            | 1.87(2)                |
| 1.4       | 27.5      | 0.398(3)            | 2.8101(1)            | 2.39(2)                | 2.4234(1)            | 3.9323(4)            | 1.86(2)                |
| 1.5       | 30.4      | 0.291(3)            | 2.8006(1)            | 2.94(3)                | 2.4165(1)            | 3.9188(3)            | 2.04(2)                |
| 2.0       | 31.3      | 0.257(3)            | 2.8390(1)            | 3.45(3)                | 2.4145(1)            | 3.9141(3)            | 2.15(2)                |
| 2.5       | 35.5      | 0.079(2)            |                      |                        | 2.4075(1)            | 3.8883(2)            | 3.29(2)                |
| 4.0       | 40.2      | 0.083(2)            |                      |                        | 2.3960(1)            | 3.8744(2)            | 3.57(2)                |
| 4.7       | 45.5      | 0.061(2)            |                      |                        | 2.3864(1)            | 3.8523(2)            | 3.92(2)                |
| 5.5       | 50.8      | 0.046(3)            |                      |                        | 2.3773(1)            | 3.8318(2)            | 4.31(2)                |
| 16.8      | 52.2      | 0.047(2)            |                      |                        | 2.3745(1)            | 3.8287(2)            | 4.22(2)                |

Fe-4.3wt%Si (Dubrovinsky *et al.* 2003). In the latter case the sample recovered from 22 GPa at 2473 K was found to be homogenous. Our sample recovered after Run#2 shows no variation of chemical composition (Figs 7e and f). Therefore, we consider chemical composition of the alloy to be uniform and constant throughout the experiment. Consequently, the observed crystalline phases are considered Fe-9wt%Si at all experimental conditions.

Many DAC experiments (e.g. Lin *et al.* 2002; Dubrovinsky *et al.* 2003; Lin *et al.* 2003; Hirao *et al.* 2004) suggest that Fe–Si alloys with Si content below  $\sim 10$  wt per cent should have a bcc structure at temperatures and pressures close to ambient conditions. Fischer *et al.* (2013; 2014) suggested that Fe-9wt%Si alloy could have a D0<sub>3</sub> structure at ambient temperature and pressures up to 29 GPa. It is reported that both B2 and D0<sub>3</sub> ordered structures may form in low-silicon Fe–Si alloys depending on material synthesis and heat treatment (Shin *et al.* 2005).

In our experiments, the only occurrence of subsidiary diffractions, indicating a certain degree of ordering of the bcc phase, is observed in Run #2 at the lowest pressure of 2.7 GPa (Fig. S1), which could be due to presence of either D0<sub>3</sub> or a B2 phase (or both). This ambiguity about the crystal structure of Fe-9wt%Si alloy at ambient temperature and low pressure cannot be resolved based on our data.

Changes of the bcc unit cell volume with pressure are comparable to other observations during compression of Fe–Si alloys with similar composition (Fig. 4a). The refined value  $V_0 = 23.18(4) \text{ Å}^3$  is within error margins from values reported by Lin *et al.* (2003) and Fischer *et al.* (2014) (the latter is for D0<sub>3</sub> Fe-9wt%Si taking into account the twice larger unit cell parameter), and are slightly higher than values obtained by Hirao *et al.* (2004). The bcc phase bulk modulus value  $K_0 = 205(8) \text{ GPa}$  is also in good agreement with previously reported values for bcc Fe–Si alloys (Fischer *et al.* 2014, their table 1).

The first appearance of the hcp phase is observed at 17 GPa during Run #1, and is already a significant fraction (33.5 volume percent). In Run #2 hcp initiates at 15.3 GPa and becomes significant at 22.3 GPa (35.7 vol percent; Table 2). The onset of the bcc  $\rightarrow$  hcp transformation in Fe-9wt%Si at  $\sim 15$  GPa, is consistent with values obtained for Fe–Si alloys with similar silicon content (Dubrovinsky *et al.* 2003; Lin *et al.* 2003; Hirao *et al.* 2004). In Run #1, the bcc phase remains in the sample up to 40.2 GPa, and in Run #2, the bcc phase is observed up to  $> 50$  GPa, though as minor quantities ( $< 6$  vol per cent). Most likely the expansion of the pressure range of bcc-hcp coexistence is due to pressure/stress gradients in the DAC, because of the absence of a pressure medium in rDAC experiments which would reduce axial stress and reduce texture development.

Interactions between crystallites (Barton *et al.* 2005) or structural defects (Von Bargen & Boehler 1990) may also expand the transition range.

The pressure dependence of the unit cell volume of hcp Fe-9wt%Si is shown in Fig. 4(b). There is a close correspondence to Fe-Si alloys with similar silicon content (Lin *et al.* 2003; Hirao *et al.* 2004; Tateno *et al.* 2015, their second run). Refined reference unit cell volume  $V_0 = 22.53(10) \text{ \AA}^3$  and bulk modulus  $K_0 = 165(7) \text{ GPa}$  are within the error margins from the results reported by Tateno *et al.* (2015).

An important crystallographic parameter for the hcp phase is the ratio  $c/a$  that may be related to texture formation mechanisms (Wang & Huang 2003; Britton *et al.* 2015). Our results show that in hcp Fe-9wt%Si in a pressure range  $\approx 25\text{--}54 \text{ GPa}$  it is about 1.62 (Fig. 4d), which is comparable to other research (e.g. Mao *et al.* 1967; Huang *et al.* 1987; Tateno *et al.* 2010; Merkel *et al.* 2020). At pressures  $\approx 15\text{--}25 \text{ GPa}$ , the  $c/a$  ratio in hcp Fe-9wt%Si is 1.63–1.64, close to the  $c/a$  ratio for ideal hcp close-packing ( $c/a = \sqrt{8/3} \approx 1.633$ ). An increased  $c/a$  ratio at the onset of the bcc  $\rightarrow$  hcp transition was observed in some experiments in pure iron (Bassett & Huang 1987; Huang *et al.* 1987; Wang & Ingalls 1998), where it was suggested to be caused by the realization of interfacial compatibility of bcc grains and hcp nuclei, though more recent results attribute this effect to differential stresses in the DAC (Merkel *et al.* 2020).

## 4.2 Texture development in bcc Fe-9wt%Si alloy

Initially Fe-9wt%Si samples had irregular CPOs, and smooth fibre CPOs formed in the bcc phase with increase of pressure and deviatoric stresses.

The first development of a bcc fibre texture was observed in Run #1 at a pressure of 4.3 GPa and corresponding  $t = 2.48 \text{ GPa}$ , and in Run #2 at a pressure of 7.6 GPa and  $t = 2.41 \text{ GPa}$  (Fig. 6c). We conclude that von Mises stress for bcc Fe-9wt%Si is  $\approx 2.4 \text{ GPa}$ , which is well below the reported  $t \approx 5 \text{ GPa}$  in pure bcc iron at pressures up to 13 GPa at ambient temperature (Merkel *et al.* 2013).

Deformation of bcc Fe-9wt%Si produces a two-component (111)+(100) fibre texture (Figs 5a and f), with (111) being the major component. This is the typical axial compression texture of bcc metals and alloys (e.g. Rollett & Wright 1998; Weinberger *et al.* 2013). It was observed in high pressure synchrotron diffraction experiments during deformation of pure bcc iron at ambient and increased temperatures, and the ratio of peak intensities of (111) and (100) components in IPFs was reported to be  $\approx 1\text{--}3$  (Miyagi *et al.* 2008; Merkel *et al.* 2013; Nishihara *et al.* 2018). This is comparable to our results for single phase bcc Fe-9wt%Si, where this ratio is  $\approx 1.7\text{--}3.2$  and increases with pressure (Fig. 6a).

Note that this 2-component bcc CPO has not only two strong texture components (at  $111 \approx 6\text{--}9 \text{ m.r.d.}$  and at  $100 \approx 2\text{--}4 \text{ m.r.d.}$ ), but also a significant fraction of randomly oriented crystallites (minimum pole density values are  $0.2\text{--}0.3 \text{ m.r.d.}$ ; Figs 5a–c, f–i). Nevertheless, observed bcc phase CPOs are much sharper than reported for pure iron (Merkel *et al.* 2004; Miyagi *et al.* 2008; 2013; Nishihara *et al.* 2018) and bcc Fe–Ni–Si alloys (Brennan *et al.* 2021).

## 4.3 Transformation texture in hcp Fe-9wt%Si

When the bcc  $\rightarrow$  hcp transformation is initiated in Fe-9wt%Si by increasing pressure, the hcp phase already has a distinct preferred orientation with (11 $\bar{2}$ 0) planes normals oriented subparallel to compression (Figs 5b and g). This is consistent with the DAC and D-DIA

compression experiments on pure iron and Fe–Ni–Si alloys, where the hcp phase formed from the bcc phase with a two-component (111)+(100) texture (e.g. Merkel *et al.* 2004; Nishihara *et al.* 2018; Brennan *et al.* 2021).

In other experiments, a different preferred orientation pattern of the newly formed hcp iron phase was observed, for example the hcp iron phase with the broad (10 $\bar{1}$ 0) maximum in the compression direction (Miyagi *et al.* 2008, their fig. 4). Nishiyama *et al.* (2007) found that just after transformation from the bcc phase at 14.5 GPa pressure and 700 K temperature hcp iron has pronounced preferred orientation with [0001] directions subparallel to the compression. But their sample was initially an iron wire with a [110] fibre texture of the bcc phase, which could not change to (111)+(100) compression texture under nearly hydrostatic loading conditions.

The transformation texture will be convoluted with the preferred orientations patterns due to plastic deformation of the hcp phase. Thus, it is important to overview the influence of the phase transition and bcc texture on the resulting CPO of hcp Fe-9wt%Si.

## 4.4 Modelling the transformation texture with variant selection

For the calculation of the transformation texture we follow a similar procedure as Yue *et al.* (2019) used for  $\text{Mn}_2\text{O}_3$ . The experimental ODF calculated with the E-WIMV method is an array of ODF values  $f(g)$  corresponding to different orientations  $g$ . Every orientation is defined by three Euler angles in Roe-Matthies convention,  $g = \{\alpha, \beta, \gamma\}$  (Matthies *et al.* 1987, their fig. 1.3), necessary to bring the sample and crystal coordinate systems (Cartesian, right-handed) to coincidence by three consecutive rotations. In E-WIMV, these angles take discrete values in  $5^\circ$  increments. Let us denote orientations in the bcc phase as  $g^{\text{bcc}}$ , and orientations in the hcp phase as  $g^{\text{hcp}}$ . Then, if there is a set of rotations, which brings in the coincidence bcc and hcp crystal coordinate systems, the orientation in the hcp phase, after the bcc  $\rightarrow$  hcp transition, may be calculated as:

$$g^{\text{hcp}} = g^{\text{HCPsym}} g^{\text{OR}} g^{\text{BCCsym}} g^{\text{BCC}} g^{\text{SAMPLE}}, \quad (1)$$

where  $g^{\text{SAMPLE}}$  corresponds to equivalent orientations due to symmetry of the sample (fibre),  $g^{\text{BCCsym}}$  are equivalent orientations due to rotational symmetry of the bcc crystal,  $g^{\text{OR}}$  is the crystallographic orientation relationship (OR) of the hcp phase relative to the parent bcc phase, and  $g^{\text{HCPsym}}$  are equivalent orientations due to rotational symmetry of the hcp crystal. Every ODF value in the bcc phase  $f(g^{\text{bcc}})$  is attributed to the new orientation  $g^{\text{hcp}}$  calculated with (1). This new function is interpolated into the discrete  $5^\circ$  grid and normalized to unity to comply with the E-WIMV formulation and ODF properties, effectively providing the hcp phase ODF.

The latter three factors of eq. (1), that is  $g^{\text{BCCsym}}$ ,  $g^{\text{BCC}}$ ,  $g^{\text{SAMPLE}}$  are already considered in the experimental ODF.

Next,  $g^{\text{OR}}$  should be defined from the crystallography of the bcc  $\rightarrow$  hcp transformation. It was first explored on single crystals of zirconium subjected to heating and Burgers (1934) suggested an OR with  $[110]_{\text{bcc}}$  parallel to  $[0001]_{\text{hcp}}$  and  $[\bar{1}11]_{\text{bcc}}$  parallel to  $[\bar{1}2\bar{1}0]_{\text{hcp}}$ . Corresponding atomic rearrangement is schematically shown in Fig. 8(a). Burgers OR was studied in many polycrystalline materials and considered for the CPO formation in hcp iron at high pressure (e.g. Merkel *et al.* 2004; Miyagi *et al.* 2008; 2013; Nishihara *et al.* 2018; 2020). Later Pitsch & Schrader (1958) discovered another OR in iron carbides that is slightly rotated from Burgers OR and this was taken into account for the bcc  $\rightarrow$  hcp transition in pure iron by Mao

*et al.* (1967) and Basset & Huang (1987). There are other possible ORs, close to Burgers OR, which include, for example Potter OR that has an additional  $\approx 2^\circ$  rotation (Potter 1973), or multiple 'near Burgers' ORs (Dahmen 1982; Zheng *et al.* 2009).

In the experiment on polycrystals with limited angular resolution of ODFs, it is impossible to distinguish between these ORs based on texture only. Therefore, we follow up on Merkel *et al.* (2020) and use Burgers OR for the description of the transformation effect on CPO of Fe-9wt%Si.

We use the following formulation for the Burgers OR:

$$\begin{aligned}[110]_{\text{bcc}} &\rightarrow [0001]_{\text{hcp}} \\ [\bar{1}\bar{1}\bar{2}]_{\text{bcc}} &\rightarrow [10\bar{1}0]_{\text{hcp}} \\ [\bar{1}\bar{1}1]_{\text{bcc}} &\rightarrow [\bar{1}2\bar{1}0]_{\text{hcp}}\end{aligned}$$

Correspondingly the three Euler angles for the bcc  $\rightarrow$  hcp crystal coordinate system rotation are  $\alpha^{\text{OR}} = 45^\circ$ ,  $\beta^{\text{OR}} = 90^\circ$ ,  $\gamma^{\text{OR}} = 5.26^\circ$  (Fig. 8b). The cubic [010] axis ( $Y_{\text{bcc}}$ ) is tilted  $45^\circ$  around the cubic [001] axis ( $Z_{\text{bcc}}$ ) into the basal hexagonal plane, afterwards cubic [001] axis is rotated  $90^\circ$  around new [010] direction and finally there is a  $5.26^\circ$  rotation around the [0001] ( $Z_{\text{hcp}}$ ). The angles do not depend on unit cell parameters, and  $\gamma^{\text{OR}} = 5.26^\circ$  here is a rounded value; the tangent of  $\gamma^{\text{OR}}$  is expressed in real radicals as  $1/(4\sqrt{2} + 3\sqrt{3})$ .

All  $g^{\text{HCPsym}}$  rotations have to be applied to comply with the crystal symmetry in the ODF. The hcp phase is attributed to the point group 6/m 2/m 2/m with 12 symmetry equivalent rotations (e.g. Matthies *et al.* 1987, their table 5.5).

The result of applying these operations to the experimental bcc phase ODF measured in Run #1 at 10.4 GPa (Fig. 5a) is shown in Fig. 9(a).

Compared to experimental CPO of the hcp phase measured in the beginning of the bcc  $\rightarrow$  hcp transformation (Figs 5b, g and h), this model hcp transformation texture has a relatively intensive component near (11 $\bar{2}$ 4) in addition to the experimentally observed maximum near (11 $\bar{2}$ 0) (Figs 9a and b). Its absence in the experiment means that a certain variant selection mechanism should be active during the bcc  $\rightarrow$  hcp transformation in Fe-9wt%Si compressed in DAC at ambient temperature.

Another observed peculiarity of Fe-9wt%Si CPOs is that when the material becomes a two-phase bcc-hcp mixture, the (100) component of the two-component bcc fibre texture is depleted (Figs 6a, 5b, c, g and h). It suggests that crystallites of the bcc phase with [100] directions close to the compression direction transform into hcp first. Another possibility is that the depletion of (100) is simply due to an increase in (111) due to further deformation.

To investigate these effects in more detail, it is necessary to explore possible variant selection, which requires investigation of different  $g^{\text{BCCsym}}$  rotations in eq. (1). The experimental ODF already considers all 24 symmetry equivalent rotations (e.g. Matthies *et al.* 1987, their table 5.2) of the bcc Fe-9wt%Si, prescribed by its point group 4/m  $\bar{3}$  2/m. To separate them we constructed model CPOs based on the experimental bcc ODF measured in Run #1 at 10.4 GPa (Fig. 5a) in Beartex (with STOD). The model bcc ODFs feature two Gaussian fibres with FWHM of  $20^\circ$  and a 0.2 m.r.d. random texture component. The relative intensity of fibres may be chosen so that the ratio of (111) and (100) maxima in compression direction after the random CPO component subtraction is 2.1, the same as in the experimental CPO (Fig. 6a). The models were made with and without equivalent rotations of the bcc structure.

The model made with account for the bcc crystal symmetry is in excellent agreement with the experimental bcc ODF (*cf.* Figs 9a and b). Model ODFs constructed without bcc crystal symmetry

were used to investigate the effect of variant selection, assuming that crystallites with orientations belonging to the particular fibre undergo a bcc  $\rightarrow$  hcp transition with a single transformation variant. For this purpose, for each of bcc fibres, (100) and (111), 24 symmetry equivalent variants were modeled and separately transformed into the hcp by applying the Burgers OR. After applying  $g^{\text{HCPsym}}$  rotations to every variant, the IPFs were calculated and grouped according to orientation distribution patterns.

For transformation of (100) bcc fibre texture into the hcp, there are only 2 distinct types of IPFs (Fig. 9c). Type I(100) IPF has a maximum at  $45^\circ$  to [0001] in direction towards (01 $\bar{1}$ 0) normal, close to (01 $\bar{1}$ 2) plane normal. The maximum is created by two closely located texture fibres. Type II(100) IPF has a broad maximum at  $90^\circ$  to (0001), but the fibre axis is not aligned with the (11 $\bar{2}$ 0) normal, it is inclined by  $5.26^\circ$  (Figs 8a, b and 9c). IPFs of type I(100) are twice more frequent (labeled  $\times 2$  in Fig. 9) compared to type II(100) IPFs: 16 versus 8 variants out of 24 total. Without variant selection, features of both I(100) and II(100) types are present in the hcp phase IPF of compression direction with comparable maximum values (4.0 and 3.6 m.r.d. correspondingly; Fig. 9c).

The transformation of the (111) bcc fibre results in three different types of IPFs (Fig. 9d). Type I(111) IPF shows a maximum at  $35.26^\circ$  to [0001] in direction towards the (11 $\bar{2}$ 0) normal. Type II(111) IPF has an intensive maximum exactly aligned with (11 $\bar{2}$ 0) normal. Type III(111) IPF is characterized by a broad maximum at  $90^\circ$  to [0001], centred close to (21 $\bar{3}$ 0). IPFs of type I(111) are twice more frequent compared to other two types: 12 versus 6 versus 6 variants. Without variant selection, bcc  $\rightarrow$  hcp transformation produces IPF with a strong (11 $\bar{2}$ 0) maximum (4.8 m.r.d.) and a weaker component brought in by type I(111) variants (3.1 m.r.d., Fig. 9d).

These simple considerations of transformation texture formation during the bcc  $\rightarrow$  hcp transition in polycrystalline Fe-9wt%Si lead to important conclusions.

First, transformation of a typical bcc compression texture does not yield preferred hcp orientations with a (10 $\bar{1}$ 0) or a (0001) maximum in the hcp phase. If such a CPO is observed right after the transition to hcp phase (e.g. Miyagi *et al.* 2008, their fig. 4), it may be because of the complexity of the transformation that gradually happens over a wide pressure range and ongoing plastic deformation, or a non-Burgers OR between bcc and hcp phases, which is unlikely.

Second, the modelling demonstrates that through a variant selection mechanism the newly formed hcp phase may obtain preferred orientation with a broad (11 $\bar{2}$ 0) IPF maximum in compression, without any subsidiary orientation components, similar to experimentally observed Fe-9wt%Si textures (Figs 5b, g and h). This is possible in the case when the phase transition first occurs in bcc crystallites oriented with [100] close to compression, with transformation realized via variants of type II(100) (Fig. 9c). In general, our data suggest that the variants with [110] bcc direction (which becomes [0001] axis in the hcp phase) aligned perpendicular to compression axis are preferred during the initial stage of bcc  $\rightarrow$  hcp transition in Fe-9wt%Si under uniaxial compression at ambient temperature.

A single component (11 $\bar{2}$ 0) hcp transformation texture, which formed from a two-component (111)+(100) bcc fibre, was previously reported for pure iron only by Merkel *et al.* (2004), while for example Nishihara *et al.* (2018) clearly observed an additional component of the hcp preferred orientation. Some Fe-Ni-Si alloys also demonstrate preferred orientation with (11 $\bar{2}$ 0) normal to compression after the bcc  $\rightarrow$  hcp transition, but Fe-5Ni-10Si CPO suggests a transformation without variant selection (Brennan *et al.* 2021). Finite elements modelling of the transformation texture (Barton *et al.*

**Table 3.** Deformation systems and CRSS values used in VPSC models.

| Deformation system                                                                                                    | CRSS |
|-----------------------------------------------------------------------------------------------------------------------|------|
| Bcc                                                                                                                   |      |
| Slip $\{110\}\langle 111\rangle$                                                                                      | 1    |
| Slip $\{110\}\langle \bar{1}10\rangle$                                                                                | 5    |
| Hcp                                                                                                                   |      |
| slip $(0001)\langle 11\bar{2}0\rangle$                                                                                | 1    |
| Slip $\{10\bar{1}0\}\langle \bar{1}2\bar{1}0\rangle$                                                                  | 4    |
| Twinning $\{10\bar{1}2\}\langle \bar{1}011\rangle$                                                                    | 2    |
| Twinning $\{2\bar{1}\bar{1}2\}, \langle 2\bar{1}\bar{1}-3\rangle, \dots-3$ use bar over 3 as for -1 in front of it... | 5    |

2005) supports weak, if any, variant selection during the bcc→hcp transformation in iron. On the other hand, recent near hydrostatic DAC experiments (Merkel *et al.* 2020) show that the bcc→hcp transition in textured pure iron is heavily affected by stress-induced variant selection, with  $[0001]$  hcp axes concentrating perpendicular to compression with minimal spread due to interactions between neighbouring grains. This supports our interpretation of the Fe-9wt%Si transformation texture, but we acknowledge that the matter of variant selection during the bcc→hcp transition in iron and Fe-Si alloys under stress is complex and requires further modelling and experiments to be clearly understood.

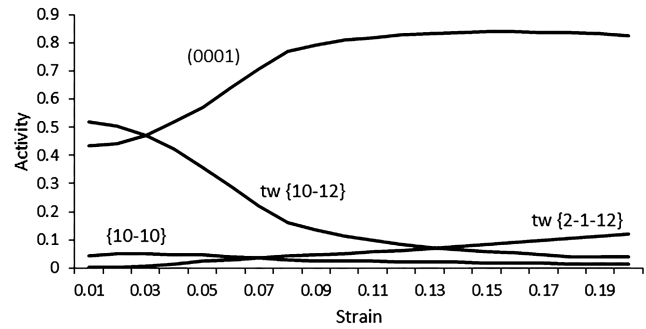
#### 4.4 VPSC modelling of texture development during plastic deformation

In the previous section we explored texture changes during the bcc-hcp phase transition quantitatively with variant selection models. Here we focus on plastic deformation mechanisms that produce rotation of crystals and thus CPO during straining.

We used the Visco-Plastic Self-Consistent (VPSC) approach (Lebensohn & Tomé 1994) that relies on activation of slip systems and mechanical twinning of crystals embedded in an anisotropic viscous medium. This model neglects elasticity as in elastoplastic models (e.g. Lebensohn *et al.* 2012; Bhattacharyya *et al.* 2021), but has no significant effect on texture development at conditions of our DAC experiments. We started with 2000 orientations, randomly oriented for bcc and for hcp with an experimentally observed  $(11\bar{2}0)$  CPO (Run #1, 17 GPa, Fig. 5b), generated by adding proper ODF weights to all random orientations.

For crystal deformation systems we assumed slip and mechanical twinning (Table 3) and plastic deformation by a power law with  $n = 5$  as stress exponent. Each assumed slip system was assigned a relative critical resolved shear stress (CRSS) to differentiate their activities. CRSS values were chosen so that calculated CPOs best fit with observed data. The aggregate was deformed in axial compression and 1 percent strain steps to 50 percent linear strain for bcc and 20 percent for hcp. CPO results are displayed in inverse pole figures (Fig. 10) and activities of deformation systems in hcp as function of strain (Fig. 11).

For the bcc phase the most common  $\{110\}\langle 111\rangle$  slip was assumed (Opinsky & Smoluchowski 1951; Watanabe 2006), producing the main (111) compression IPF maximum of 10 m.r.d. and a secondary maximum at (001) of 7.2 m.r.d. (Fig. 10a), corresponding to a typical bcc compression texture (Rollet & Wright 1998) and closely resembling experimentally observed bcc CPO (Figs 5a–c, f–i). 50 % strain provided the best fit with observed data. While



**Figure 11.** VPSC model of hcp iron deformed in axial compression. Activities of slip and mechanical twinning systems (in fractions) as function of strain.

this seems rather high it is not unreasonable for the initial sample compression in the DAC.

The principal slip system of hcp metals with a  $c/a$  ratio close or higher than  $\approx 1.633$  is basal slip (e.g. Partridge 1967; Wang & Huang 2003; Britton *et al.* 2015). Additionally, prismatic slip was found to be moderately active in pure hcp iron (Merkel *et al.* 2004; Miyagi *et al.* 2008). Therefore, for the Fe-9wt%Si hcp phase, dominant basal slip  $(0001)\{11\bar{2}0\}$  (CRSS = 1.0) and secondary prismatic slip  $\{10\bar{1}0\}\langle \bar{1}2\bar{1}0\rangle$  (CRSS = 4.0) were used in the model. In addition mechanical twinning was included, both  $\{10\bar{1}2\}\langle \bar{1}011\rangle$  and  $\{2\bar{1}\bar{1}2\}\langle 2\bar{1}\bar{1}-3\rangle$ , which is an important mechanism in hcp metals (e.g. Partridge 1967; Yoo 1981; Kanitpanyacharoen *et al.* 2012; Liu *et al.* 2021).

We start hcp VPSC with a  $(11\bar{2}0)$  maximum (6.2 m.r.d.) produced by the bcc-hcp phase transition (Fig. 10b). In this CPO (0001) is at right angle to the compression and basal slip is not active, except for broad background orientations (the minimum is only 0.3 m.r.d.). In the first few steps  $\{10\bar{1}2\}\langle \bar{1}011\rangle$  twinning dominates (0.52), basal slip is nevertheless active (0.43) and there is minor prismatic slip (0.05) (Fig. 11). After 10 steps (10 % strain) twinning is reduced to 0.1 and basal slip has increased to 0.8. In terms of textures, the  $(11\bar{2}0)$  maximum is reduced from 6.2 m.r.d. (Fig. 10b) after 5 steps to 5.7 m.r.d. (Fig. 10c) and has largely disappeared after 10 steps (Fig. 10d). Correspondingly the maximum near (0001) has increased to 7.9 m.r.d. after 10 steps and reaches 11.0 m.r.d. after 20 steps (Fig. 10e), with a similar pattern as observed experimentally in Run #2 (Figs 5k–n).

#### 4.5 Texture development in hcp Fe-9wt%Si alloy

Due to variant selection during the bcc→hcp transformation, a strong transformation texture is formed in hcp Fe-9wt%Si with a maximum at  $(11\bar{2}0)$  (Figs 5b and g). During Run #1, the initial hcp phase transition induced  $(11\bar{2}0)$  IPF component gradually decreases in strength with the increase of the hcp phase content. An additional IPF maximum near  $(11\bar{2}4)$  may be related to transformation with  $I(111)$  variants (Figs 5c and d). The increase in pole density closer to (0001) (Fig. 5d) is due to plastic deformation.

In Run #2 after laser heating, hcp becomes the dominant phase ( $\sim 95$  per cent) and followed by further compression the  $(11\bar{2}0)$  disappears and the (0001) maximum dominates (3.5–3.9 m.r.d., Figs 5j and k).

In the VPSC model of the hcp phase deformation texture, basal slip  $(0001)\langle 11\bar{2}0\rangle$  was assumed as the dominant slip system (Table 3). There are many randomly oriented grains in the transformation texture (e.g. Fig. 10b, minimum 0.3 m.r.d.) and they are

susceptible to basal slip. But grains at the (11 $\bar{2}$ 0) maximum are at 90° to (0001) and cannot deform by basal slip. They are reoriented early by {10 $\bar{2}$ 1} twinning (Fig. 11) that quickly depletes the (11 $\bar{2}$ 0) maximum (Figs 10c–e).

In pure iron observed differential stresses are significantly reduced at high temperatures (Nishiyama *et al.* 2007; Merkel *et al.* 2012, 2013; Nishihara *et al.* 2018). Therefore it is possible that slow development of the hcp deformation texture in Run#1 and after laser heating in Run#2 is because differential stress increments are small at high pressures with diamonds in close contact, but during the laser heating critical stresses in Fe-9wt%Si are also reduced, and large plastic deformation is possible.

In terms of texture sharpness, the deformation hcp Fe-9wt%Si CPO is comparable to the majority of reported compression textures of pure hcp iron and Fe–Ni–Si alloys at similar pressures, where maximum pole density values in IPFs are  $\sim 2$ –3 m.r.d. (Merkel *et al.* 2004; Mao *et al.* 2008; Miyagi *et al.* 2008, 2013; Nishihara *et al.* 2018; Brennan *et al.* 2021).

#### 4.6 Preferred orientations on decompression and general remarks

Some data were obtained on preferred orientations of Fe-9wt%Si during unloading. Unfortunately, no conclusive data for the inverse hcp  $\rightarrow$  bcc transition were obtained because pressure was not completely released while the DAC cell was in the X-ray beam, and only a small amount of the hcp phase transformed back to bcc. IPFs of bcc Fe-9wt%Si at reduced pressure show a significant but weaker (111) maximum (Figs 5e and n), and during decompression in Run #1, a weak (100) maximum starts to form (Fig. 5e). Decompression IPFs of the hcp phase are quite similar to high temperature tension deformation textures observed in pure iron (Nishihara *et al.* 2018).

It is tempting to extrapolate the results to macroscopic deformation models at pressures, temperatures and strain rates of the inner core to assess seismic anisotropy. However, as advised by results of radial DAC and D-DIA deformation experiments on iron (e.g. Merkel *et al.* 2013; Nishihara *et al.* 2018), different deformation mechanisms may be active at increasingly higher temperatures. Temperature could have a profound impact on deformation CPOs of Fe–Si alloy at high pressures.

The rDAC experiments clearly have limitations. There are significant pressure and stress gradients between the centre and the edge of the sample and in radial geometry the beam averages over both. This is a likely reason why bcc and hcp seem to coexist over a wide pressure range. Another limitation is that samples are very thin (30–40  $\mu$ m after initial compaction). This limits the amount of strain that can be imposed, especially at higher pressures when diamonds are in close contact. It may explain why in run #2 nothing seemed to change significantly when pressures were raised from 31 to 52 GPa over long times. This is particularly significant for the bcc–hcp mixture of iron alloys, where much of the deformation may occur in the bcc phase. Here D-DIA experiments provide better control over pressure, stress and strain but the pressure range is limited. Theoretical modelling of slip systems activity in hcp materials that rely on elastic properties and stacking fault energies (e.g. Poirier & Price 1999) open new opportunities.

## CONCLUSIONS

Synchrotron X-ray diffraction experiments on a Fe-9wt%Si alloy with DAC in radial geometry documented phase transformations, changes of unit cell parameters and crystal preferred orientations at pressures up to 54.1 GPa. A characteristic two component (100)+(111) compression texture was observed for bcc Fe-9wt%Si, indicating dominant {110}<111> slip. It is similar to preferred orientations observed in pure bcc iron. A transformation texture is formed in the hcp phase during the initial stage of the bcc  $\rightarrow$  hcp transition with {11 $\bar{2}$ 0} planes oriented perpendicular to compression. It is described by Burgers orientation relationship with variant selection preferring those with [110] bcc/[0001] hcp directions perpendicular to the compression axis. Analysis of preferred orientations suggests that bcc grains oriented with [100] parallel to compression transform to the hcp phase first. The hcp transformation texture and high yield strength of hcp Fe-9wt%Si hinders plastic deformation at ambient temperature. After heating the alloy to 1600 K, the formation of a deformation texture was observed. The observed compression texture of hcp Fe-9wt%Si is characterized by alignment of (0001) planes perpendicular to compression that can be interpreted as dominant (0001)<11 $\bar{2}$ 0> slip. It also involves mechanical twinning. Deformation experiments at higher temperatures and pressures, preferably close to inner core conditions, combined with first principle calculations on Fe-9wt%Si should provide more information for texture formation mechanisms in this material, but our results suggest that Fe–Si alloys have similar slip systems as pure iron at high pressure.

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## DATA AVAILABILITY

The data in this paper, including diffraction images, will be shared on reasonable request to the corresponding author.

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