EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

# Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF**.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

*https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do*

***Reports supporting requests for additional beam time***

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

***Published papers***

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

## Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

## Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

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|---|---|---|
| ESRF | **Experiment title:**<br>***In situ high PT study of the micromechanical properties of the major Earth`s mantle constituent $(Mg,Fe)_2SiO_4$ during its phase transformations, implications for geodynamics.*** | **Experiment number**:<br>ES-98 |
| **Beamline**:<br>ID27 | **Date of experiment**:<br>from: 9th April 2014 to: 15th April 2014 | **Date of report**:<br>21st July 2014 |
| **Shifts:**<br>18 | **Local contact(s)**:<br>Gaston Garbarino | *Received at ESRF:* |


**Names and affiliations of applicants** (* indicates experimentalists):

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## ***Report:***

The series of phase transitions in the dominant upper mantle constituent $Mg_{1.9}Fe_{0.1}SiO_4$ [olivine α – phase] to its high-pressure polymorphs wadsleyite [β- phase] and ringwoodite [γ-phase] play an essential role for large scale geodynamical processes in the Earth's mantle (*Ringwood and Major,* 1966; *Weidner and Wang,* 2000). Several features in geophysical observations remain difficult to explain, including seismic anomalies at transition zone depth, where these transformations take place and the diversity of slabs subduction behaviors which either stagnate in the transition zone or penetrate directly into the lower mantle. In order to assess the effects of phase transformations on the rheological properties of the slab material and to build reliable models of mantle flow and slab subduction behaviors, a detailed understanding of the microstructure development, underlying transformation mechanism and transformation kinetics is needed (*i.e.*, *Nakagawa et al.,* 2009). However, few, mostly ex situ studies have been dedicated to these topics because of the manifold technical and analytical challenges to overcome. Indeed, this type of studies require the combination of advanced 3D-XRD and optimized high-pressure high temperature diamond anvil cells techniques.

## ***Technical Part:***

**During the allocated beam time at the ESRF beamline ID27, we successfully conducted simultaneous high pressure and temperature (high PT) three dimensional-X-ray diffraction (3D-XRD) experiments up to 32 GPa and 1150 K and collected X-ray diffraction (XRD) patterns during the series of phase transitions from the α-phase to the β-phase and from the β-phase to the γ-phase.** A polycrystalline sample with a hydrous Mg-end member olivine composition ($Mg_2SiO_4$) with ~500 ppm $H_2O$ synthesized at ETH Zürich served as a starting material. The X-ray beam characteristics at the ESRF beamline ID27 consisted of a 7 (H) x 4(V) $\mu m^2$ focused monochromatic X-ray beam tuned to a wavelength of 0.3738 Å. XRD patterns were collected using a PerkinElmer detector with an active area of 2048 x 2048 pixels of 200 μm located at a distance of 457 mm from the sample. A $LaB_6$ standard was measured at the beginning of the experiment in order to obtain precise calibration parameters of the beam center position, sample to detector distance and detector tilts.

Three high PT assemblies were prepared during the allocated beam time using the setup detailed in the report (HS-4765) with additional improvements (as proposed in the previous report HS-4765), **including a modified resistive heating DAC with K-type thermocouples, a larger vacuum vessel, more stable power supplies, a remote temperature controller, and thinner cylindrical graphite heaters of 0.5 mm thickness**. In all experiments the sample was loaded without pressure transmitting medium, together with gold and NaCl for pressure-temperature determination the using cross calibration method. The gold and NaCl standards were positioned at the edge of the sample chamber in order to avoid diffraction peak overlapping. This modified setup was very well adapted for the long duration of 3D-XRD experiments during the succession of the high PT phase transformations in $Mg_2SiO_4$.

## ***Results:***

3D-XRD images were collected at ambient conditions and at low pressures (4 GPa) to precisely determine and track the initial micromechanical properties of olivine. The sample was then stepwise heated up to 1150 K in 100 ° increments with a rate of 15 ° /min. Upon heating the pressure in the sample increased without further additional compression due to the thermal expansion of the gas in the DAC membrane and thinning of the metallic gasket due to enhanced plastic flow at the high-temperatures. In the later stages of the experiments when the pressure stabilized the temperature was further increased up to 1250 K (Figure 1). Upon compression and heating 3D-XRD images were collected at each PT step to monitor the micromechanical properties of the transforming material. A total of three runs has been performed and 100 PT points were sampled up 32 GPa and 1250 K (Figure 1). Among them, we could observe the α-β transformation in two experiments and in one of these experiments the sample could be further transformed from the β-phase to the γ-phase.

Data processing is currently in progress using single-grain analysis technique and the 3D-XRD software FABLE (as in *Nisr et al.*, 2012). **The collected diffraction images are of excellent quality for 3D-XRD studies (Figure 2) and will therefore allow extracting various information from individual grains inside the polycrystalline sample (crystallographic parameters, orientations and growth rates). This analysis allows drawing conclusions on the entire sample properties (grain transformation kinetics, evolution of the grain size distributions, texture relations between parent and newly formed phase, etc.) and on the underlying phase transformation mechanisms.** The new dataset will not only allow drawing a more complete picture of phase transitions in the most abundant minerals in the Earth upper mantle but will certainly shed new light on the origin of seismic anisotropies in the vicinity of deep slabs.

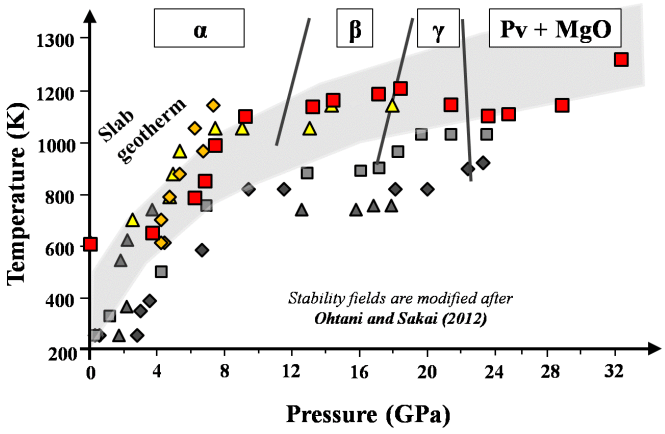


***Figure 1.*** *Pressure and temperature path ways followed during 6 different experimental runs performed during the allocated beamtime of ES-98 (colored symbols) and HS-4765 (grey symbols) plotted in the PT diagram of $Mg_2SiO_4$. Each color corresponds to a different run. The regime of slab geotherms is presented as grey shaded area while the stability fields of phases in the $Mg_2SiO_4$ system are separated by solid black lines. Squares represent selected PT points were diffraction images for 3D XRD analysis were taken. In experiments performed at temperatures below 880 K (grey diamonds and triangles) metastable persistence of the α – phase up 30 GPa was observed. In an experiments performed at higher temperatures (grey squares) the direct transformation from the α – phase to the γ-phase could be observed at 930 K and above 18 GPa. The series of phase transformations from the α- to the β- to the γ- could be monitored in a single experimental run (red squares) with increasing pressure and temperatures up to 32 GPa and 1250 K.*

## ***References:***


**Nakagawa**, T., P.J. Tackley, F. Deschamps and J.A.D. Connolly (2009), Incorporating self-consistently calculated mineral physics into thermochemical mantle convection simulations in a 3-D spherical shell and its influence on seismic anomalies in Earth's mantle, Geochem. Geophys. Geosyst., 10, *Q03004, doi:10.1029/2008GC002280.*

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**Ringwood**, A. E. and A. Major (1966), Some high pressure transformations in olivines and pyroxenes, J. Geophys. Res., 71(18), 4448.

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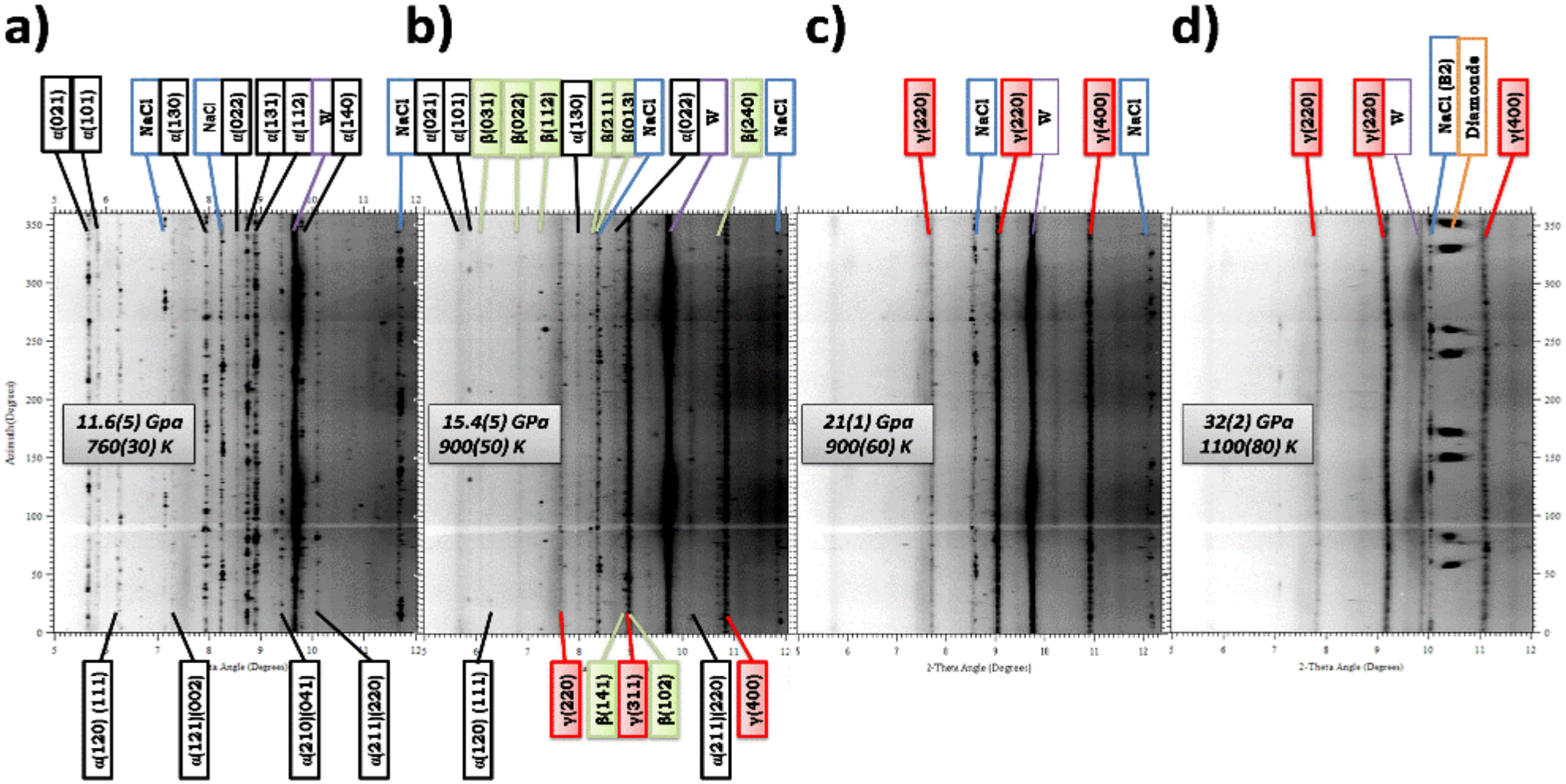


***Figure 2.*** *Unrolled diffraction patterns showing the succession of the transformation in one experimental run at four different PT points (run represented as red squares in Figure 1). Diffraction lines and Miller indices corresponding to a certain phase are indicated in black for the α–phase, in green for the β –phase and in red for the γ-phase. In* ***a)*** *the pure α–phase is stable showing a spotty diffracting signal. In* ***b)*** *the sample transformed partly to the β-phase and γ-phase, while in* ***c)*** *the sample completely transformed to the α–phase. In* ***d)*** *the γ-phase persist meta stable even though the equilibrium boundary is overstepped by more than 5 GPa. We did not observe the decomposition of the γ-phase to periclase and perovskite which might be due to the sluggish transformation kinetics. At these high pressures conditions (32 GPa) the diffraction lines of the γ-phase show a significant modulation most likely due to deviatoric stress.*