Experiment Report Form

	Experiment title: Mössbauer study of high-pressure Fe_7O_9 and Fe_9O_{11} iron oxides	**Experiment number**: HC-5325
Beamline: ID18	**Date of experiment**: from: 18.04.2023 to: 24.04.2023	**Date of report**: 30.08.2023
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Report:

This experiment aimed to investigate the high-pressure and low-temperature behavior of unexplored Fe_7O_9 and Fe_9O_{11} oxides. The Fe_7O_9 and Fe_9O_{11} samples were synthesized in Bavarian Geoinstitut (BGI) using a multi-anvil apparatus in the pressure-temperature stability field of the corresponding phases. The analysis of the synthesis products showed that Fe_7O_9 samples were crystalized as high-quality single crystals. However, the synthesis of the Fe_9O_{11} samples resulted in the mixture of Fe_4O_5 and Fe_5O_6 phases in a proportion equal to the Fe_9O_{11} stoichiometry, likely due to the high instability of the Fe_9O_{11} phase at ambient conditions and its decomposition. In this experiment, we, thus, focused on the electronic and magnetic transitions of Fe_7O_9 at high pressure and low temperature.

During the experiment, we performed the energy-domain synchrotron Mössbauer source (SMS) spectroscopy of Fe_7O_9 in membrane-driven cryogenic diamond anvil cells at ID18 up to 83 GPa and down to 5 K (Figure 1a). **We identified several magnetic and electronic transitions.**

In agreement with the literature data, Fe atoms on both octahedral and prismatic sites in Fe_7O_9 are nonmagnetic at ambient conditions (Figure 1c). Under compression up to ~ 17 GPa at room temperature or cooling down to ~ 240 K at ambient pressure, Fe on octahedral and prismatic sites both simultaneously onset a magnetic ordering (Figure 1c). Upon cooling down to ~ 80 K at ambient pressure, we identified another magnetic transition, clearly seen from the appearance of the new peak in the center of the spectra (Figure 1c: Unknown phase 1). The further compression and cooling down results in several other magnetic transitions in Fe_7O_9 (Figures 1a and c: Unknown phase 2 and Unknown phase 3). At room temperature, the spin transition in Fe_7O_9 starts at ~58 GPa and is completed at ~83 GPa, with Fe atoms on both octahedral and prismatic sites being completely in the low spin state (Figure 1c). **Our SMS experiments, thus, clearly demonstrate that Fe_7O_9 has a very complicated high-pressure low-temperature phase diagram and further investigation is needed, particularly the structural details of the new phases.**

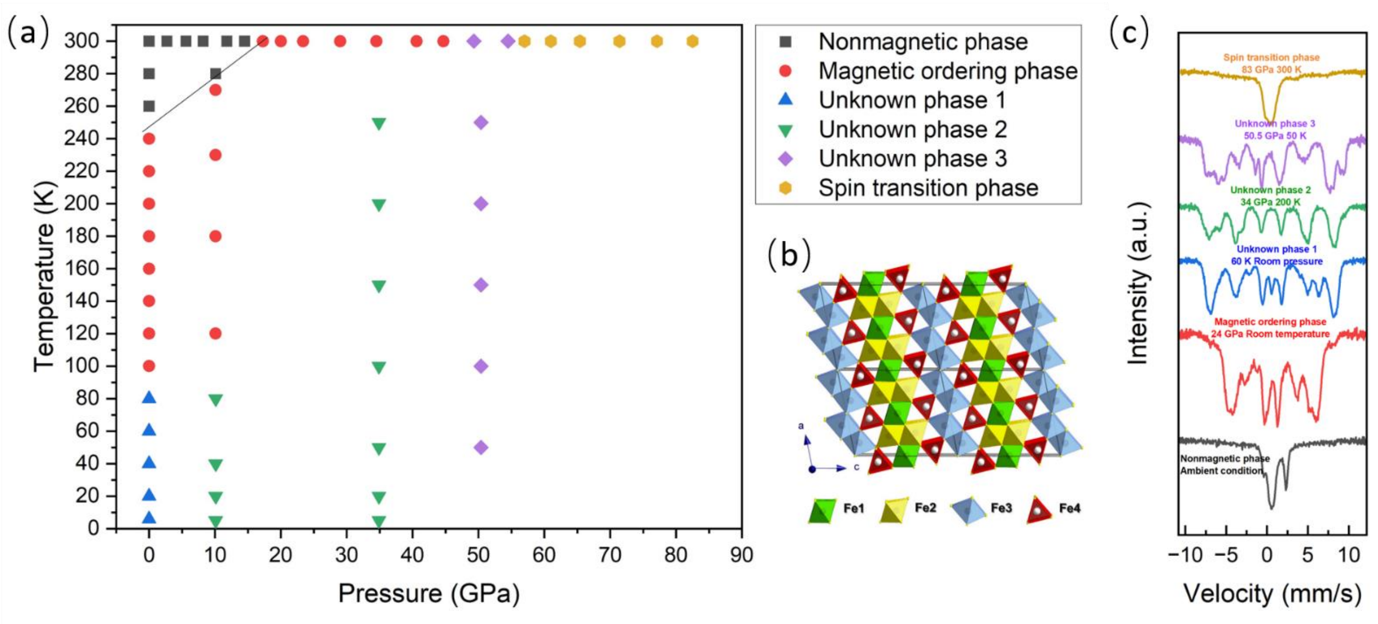

Figure 1. (a) Diagram of pressure-temperature coverage in our SMS experiments. (b) Structure of Fe_7O_9 at ambient conditions. (c) Selected SMS spectra were measured at indicated pressure and temperature conditions. The colors correspond to those in (a).