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Iron spin transitions in the lower mantle

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Iron has the ability to adopt different electronic configurations (spin states), which can significantly influence mantle properties and dynamics. It is now generally accepted as a result of studies over the past decade that ferrous iron in (Mg,Fe)O undergoes a high-spin to low-spin transition in the mid-part of the lower mantle; however results on (Mg,Fe)(Si,Al)O_3 perovskite, the dominant phase of the lower mantle, remain controversial.

Identifying spin transitions in (Mg,Fe)(Si,Al)O_3 perovskite presents a significant challenge. X-ray emission spectroscopy provides information on the bulk spin number, but cannot separate individual contributions. Nuclear forward scattering measures hyperfine interactions, but is not well suited to complex materials due to the non-uniqueness of fitting models. Energy-domain Mössbauer spectroscopy generally enables an unambiguous resolution of all hyperfine parameters which can be used to infer spin states; however high pressure measurements using conventional radioactive point sources require extremely long counting times. To solve this problem, we have developed an energy-domain synchrotron Mössbauer source that enables rapid measurement of spectra under extreme conditions (both high pressure and high temperature) with a quality generally sufficient to unambiguously deconvolute even highly complex spectra.

We have used the newly developed method to measure high quality Mössbauer spectra of different compositions of (Mg,Fe)O and (Mg,Fe)(Si,Al)O_3 perovskite at pressures up to 122 GPa and temperatures up to 2400 K. Experiments were carried out at the European Synchrotron Radiation Facility on the nuclear resonance beamline ID18 equipped with a portable laser heating system for diamond anvil cells. Our results confirm previous observations for (Mg,Fe)O that show a broad spin crossover region at high pressures and high temperatures, and show unambiguously that ferric iron in (Mg,Fe)(Si,Al)O_3 perovskite remains in the high-spin state at conditions throughout the lower mantle. Electrical conductivity data of (Mg,Fe)(Si,Al)O_3 perovskite are known to show a drop in conductivity above 50 GPa, which combined with our new results suggests that the currently controversial high-pressure transition of ferrous iron is indeed due to a high-spin to intermediate-spin transition at conditions near the top of the lower mantle. Our current picture of iron in the lower mantle is therefore of a relatively homogeneous spin state in (Mg,Fe)(Si,Al)O_3 perovskite throughout most of the lower mantle: intermediate-spin ferrous iron and high-spin ferric iron. Different spin states are expected in ferrous iron in (Mg,Fe)(Si,Al)O_3 perovskite only at the very top of the lower mantle (high spin) and at the very bottom (low spin). There is a broad transition from high-spin to low-spin ferrous iron in (Mg,Fe)O in the mid part of the lower mantle. Implications of these results for mantle properties and dynamics will be presented.