Spin transition of Fe²⁺ in ringwoodite (Mg,Fe)₂SiO₄ at high pressures

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ABSTRACT

Electronic spin transitions of iron in the Earth's mantle minerals are of great interest to deep-Earth researchers because their effects on the physical and chemical properties of mantle minerals can significantly affect our understanding of the properties of the deep planet. Here we have studied the electronic spin states of iron in ringwoodite ($Mg_{0.75}Fe_{0.25}$)₂SiO₄ using synchrotron Mössbauer spectroscopy in a diamond-anvil cell up to 82 GPa. The starting samples were analyzed extensively using transmission and scanning electron microscopes to investigate nanoscale crystal chemistry and local iron distributions. Analyses of the synchrotron Mössbauer spectra at ambient conditions reveal two non-equivalent iron species, (Fe²⁺)₁ and (Fe²⁺)₂, which can be attributed to octahedral and tetrahedral sites in the cubic spinel structure, respectively. High-pressure Mössbauer measurements show the disappearance of the hyperfine quadrupole splitting (*QS*) of the Fe²⁺ ions in both sites at approximately 45–70 GPa, indicating an electronic high-spin (HS) to low-spin (LS) transition. The spin transition exhibits a continuous crossover nature over a pressure interval of ~25 GPa, and is reversible under decompression. Our results here provide the first experimental evidence for the occurrence of the spin transition in the spinel-structured ringwoodite, a mantle olivine polymorph, at high pressures.

Keywords: Ringwoodite (Mg,Fe)₂SiO₄, high pressure, spin crossover, Mössbauer spectroscopy