AR-NE1A/2013G079

Relationship between structural variation and spin transition of hcp-Fe under high pressures and high temperatures

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1 Introduction

The high-pressure spin state of Fe is key to the understanding of magnetic, electronic, and structural properties of *3d* electrons and of the structure of the Earth's or other planets' interiors. It is known from experiments that the pressure-induced transition from body-centered cubic (bcc) to hexagonal close-packed (hcp) is accompanied by a simultaneous magnetic transition from the ferromagnetic to nonmagnetic states, because Mössbauer effect experiments have never detected the presence of a hyperfine magnetic field in the hcp structure. However, some experimental observations of the structural variation, x-ray emission spectroscopy, or Raman mode splitting indicated that hcp-Fe could have remnant magnetism. According to the structural variation, first-principles calculations predicted that the c/a ratio of the hcp structure in nonmagnetic Fe increases as the pressure increases. In contrast, recent experimental observations showed that the c/a ratio of hcp-Fe decreased with increasing pressure, indicating that hcp-Fe has the antiferromagnetic state. As the pressure increases, the magnetic transition from the antiferromagnetic to the nonmagnetic state occur, and is accompanied by spin collapse at ~60 GPa. This spin transition from the high- to the low-spin states was predicted by the first-principles calculations and was confirmed from the structural variation using the high-pressure experiments. In this study, we investigated the pressure-induced spin transition at high temperatures using the structural variation of hcp-Fe.

2 Experiment

The high-pressure and high-temperature X-ray diffraction experiments were performed using a laser-heated diamond anvil cell high-pressure apparatus. Fe powder was loaded into a 30-100 μm diameter holes drilled into a rhenium gasket, which was preindented to a thickness of 20-70 μm. The sample was sandwiched between pellets of NaCl powder to reduce any residual nonhydrostatic stresses. The sample was probed using angle-dispersive X-ray diffraction, employing the NE1A synchrotron beamline at KEK. The angle-dispersive X-ray diffraction patterns were obtained on an image plate system. The spectra were collected for about 5-10 min. The observed intensities on the imaging plates were integrated as a function of 2θ using the ESRF Fit2d software code to obtain conventional, one-dimensional diffraction profiles.

3 Results and Discussion

The structural variation in the c/a ratio of hcp-Fe was investigated up to 180 GPa and 2000 K. Prior to laser heating, we compressed the α-Fe starting material to the desired pressure at room temperature, and found that it transformed to the hcp structure. The room-temperature data were in general agreement with those reported in our previous study. The c/a ratio decreased from 1.61 to 1.60 as the pressure increased to 60 GPa, and remained approximately constant at 1.60 after that. At high temperatures, the c/a ratio decreased at 2000 K with increasing pressure. At pressures above 140 GPa, the ratio deviated noticeably from the decreasing trend. These results show that the spin transition of Fe occurred at ~140 GPa at 2000 K [1]. The gradient of dP/dT of the spin transition boundary was positive. The boundary shown in Fig. 1 is represented by the linear equation:

$$P\ (\mathrm{GPa}) = 51 + 0.045 \times T\ (\mathrm{K}).$$

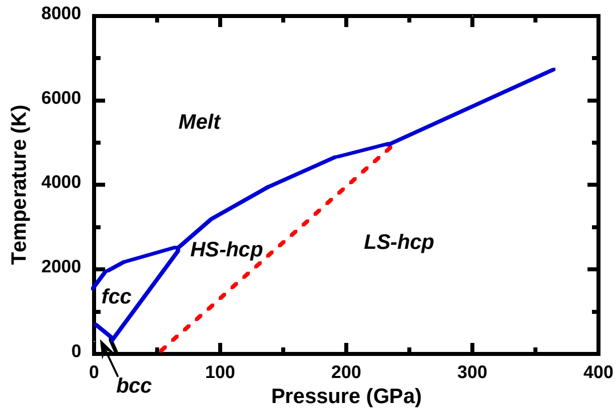

Fig. 1: Schematic phase diagram of iron at high pressures and temperatures. The dashed line represents the boundary between the high-spin (HS) and low-spin (LS) states in hcp-Fe.

Acknowledgement

We thank the PF staffs for their helps in carrying out the experiments.

References

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