13A/2005G122

Spin transition of Fe_3C

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Introduction

It is known that the Earth's inner core is less dense than pure iron, indicating that the chemical composition of the inner core is not pure iron. A significant portion of the inner core consists of light elements such as C, S, O, Si and H. Previous study proposed that the inner core might consist of Fe_3C, because the density of Fe_3C determined from its equation of state at the inner core pressure agrees with that determined from seismic observations. Cosmochemical arguments and thermodynamic calculations of the carbon-bearing iron system are also consistent with the model that proposes that the major component of the inner core is Fe_3C. Therefore, an experimental investigation of Fe_3C at extreme pressures and temperatures is important in effort to test the Fe_3C inner core model.

Recently, we have succeeded to identify the pressure-induced spin transition of iron alloys, FeO [1] and FeS [2], using X-ray diffraction technique at the synchrotron facilities. Therefore, our technique improved in previous studies was applied to the Fe_3C study. We made use of a high-pressure diamond anvil cell apparatus to investigate the physical properties of Fe_3C at high pressures. In order to assess the Fe_3C inner core model, we studied the spin transition at high pressures and examined the isothermal bulk modulus using the experimental method. We also determined the magnetic phase diagram of Fe_3C from previous studies and our new data.

Experimental

High-pressure X-ray diffraction experiments were performed using a laser-heated diamond anvil cell high-pressure apparatus. The starting material was polycrystalline Fe_3C, synthesized from pure iron and graphite powder in a platinum capsule at 1.5 GPa and 1373 K over 2.0 hours, using a piston-cylinder device. X-ray diffraction measurements, Raman spectroscopy data, and electron microprobe analyses confirmed the crystal structure and the composition of the product to be Fe_3C cementite. Fe_3C has an orthorhombic structure, space group *Pnma*, with 16 atoms in the conventional cell. The structure can be viewed in terms of a pleated layer of iron atoms, derived from a close-packed hexagonal structure with carbon atoms occupying interstitial sites. At ambient pressure and temperature, Fe_3C is metallic and ferromagnetically ordered. From the viewpoint of crystallography, Fe_3C has two iron (4c and 8d) sites and one carbon (4c) site. According to the previous Mössbauer spectroscopy data, the two different iron sites are magnetically nonequivalent. In high-pressure experiments, NaCl powder was used as an internal pressure calibrant. The samples were heated with a laser to overcome any potential kinetic effects on possible spin transitions. The samples were probed using an angle-dispersive X-ray diffraction technique at the synchrotron beam lines BL13A, Photon Factory. A monochromatic incident X-ray beam was used. The X-ray beams were collimated to a diameter of 30 μm, and the angle-dispersive X-ray diffraction patterns were obtained on an imaging plate.

Results and Discussion

In the experimental run, the sample was compressed to 15 GPa. Before laser heating was carried out, the diffraction pattern of the sample showed broad peaks, reflecting the presence of a compression-related differential stress. The sample was then heated 5~10 minutes to 1500~2500 K to relax the differential stress. During the heating stage, any new diffraction peaks did not appear, and the staring material remained stable after the temperature quench. The cell parameters and volumes were measured using X-ray diffraction patterns. Although the anisotropy of cell compression was small, a remarkable phenomenon was observed at around 55 GPa. The *a* and *c* axes decreased continuously as the pressure increased. In contrast, the *b* axis showed a discontinuous reduction at 55 GPa. The ~2% volume reduction without any structural phase transitions was observed at pressure corresponding to the discontinuous reduction of the *b* axis. We compared our data with those from previous DFT calculations. The DFT calculations indicate that the spin transition accompanied with the magnetic transition from the ferromagnetic to the paramagnetic state occurs at high pressures. Our observations of the discontinuous reductions of the *b* axis and volume were in good agreement with the spin transition predicted by the DFT calculations.

References

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