Synchrotron X-ray and Acoustic Studies of bcc-hcp Phase Transition of Fe

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Introduction

Elastic properties of hcp iron (Fe) provide crucial information about the composition and anisotropy of the Earth's core. Recently, the phonon densities of state of hcp Fe at high pressure have been investigated up to 42 and 153 GPa by using inelastic nuclear resonance x-ray scattering. The results showed discrepancies from previous results of first-principle calculations [1, 2]. The lattice strain method [3], based on measurements of stress-induced strain along various orientations, yielded information about the elastic constants of hcp Fe to pressures of >220 GPa [4]. However, some studies argue that this method requires some assumed values in the parameterization, which result in large uncertainties in the derived elastic properties [5]. Recent results on the sound velocity of Fe from using inelastic x-ray scattering [6] support previous ultrasonic velocity measurements at ~17 GPa and results from radial x-ray diffraction [4]. In this study, we measured the elastic property of hcp Fe up to 17 GPa. These data will provide critical anchor points for those ultrahigh-pressure studies as well as for theoretical calculations.

Methods and Materials

Technical advances in high-pressure ultrasonic interferometry have allowed us to measure sound wave velocities at pressure and temperature (P-T) conditions up to the top of the lower mantle [7]. The combination of ultrasonic measurements with results from a synchrotron x-ray source has enabled the simultaneous determination of the crystal structure and elasticity of a millimeter-sized solid.

In this study, we applied these state-of-art techniques to study hcp Fe by using an MA-8-type high-pressure apparatus installed at the GSECARS facility at the APS [8]. A 14/8 cell assembly modified from Ref. 9 was used in this study. In current experiments, x-ray diffraction spectra, measurements of the travel times of acoustic waves in the sample, and x-ray images of the sample were collected simultaneously at each P-T condition. Energy-dispersive x-ray diffraction spectra from the sample and an internal pressure standard (NaCl) were recorded by using a solid-state Ge detector. Ultrasonic signals were generated by using a dual-mode piezoelectric transducer made of $LiNbO_3$, and the waveform data were collected by using a recently developed transfer function method [10]. Sample images at all the P-T conditions were recorded by using a system consisting of a yttrium-aluminum-garnet (YAG) crystal and charge-coupled device (CCD) camera [8]. Sample images recorded at 13 and 17 GPa are shown in Fig. 1. Although the sample was cylindrical and sat upright in the cell, only the portions between the tungsten carbide cubes were visible to the x-ray. The real width of the sample was about the size of the whole image. The brightness contrast on these images was caused by the difference in the x-ray absorption coefficients of different materials. The materials on the top and bottom of the sample appear as white regions, while the sample appears as the dark region in the middle.

The unquenchable nature of hcp Fe prohibited us from recovering this phase at ambient conditions. Upon the release of pressure, the hcp phase reverts back to the bcc structure. In the current experiment, a 2-mm-diameter cylinder (99.9995% purity) was used as the starting material. Both ends of the cylindrical sample were polished by using 0.25-μm diamond paste.

Results and Discussion

In this experiment, the maximum pressure of ~17 GPa was reached at room temperature. Then the sample was heated to ~650K (estimated from electric power) to release the deviatoric stress caused by cold compression. For the same purpose, before data were collected at each pressure during decompression, a heating/cooling cycle was always performed. The sample pressure was determined from the x-ray diffraction of NaCl enclosed in the cell next to the sample.

X-ray diffraction data showed that the hcp phase starts to appear at ~13 GPa and continues to grow with increasing pressure. At a peak pressure of ~17 GPa, a scan of the entire sample region in Fig. 1 did not find any remaining bcc phase.

The travel-time data for compressional (P) and shear (S) waves from the current experiment are summarized in Figs. 2 and 3, respectively. As can be seen in the figures, the travel times of P and S waves decreased rapidly at a pressure range lower than 8 GPa. This could have been caused in part by the gradual shortening of the sample (as a result of the further densification of the sample at low

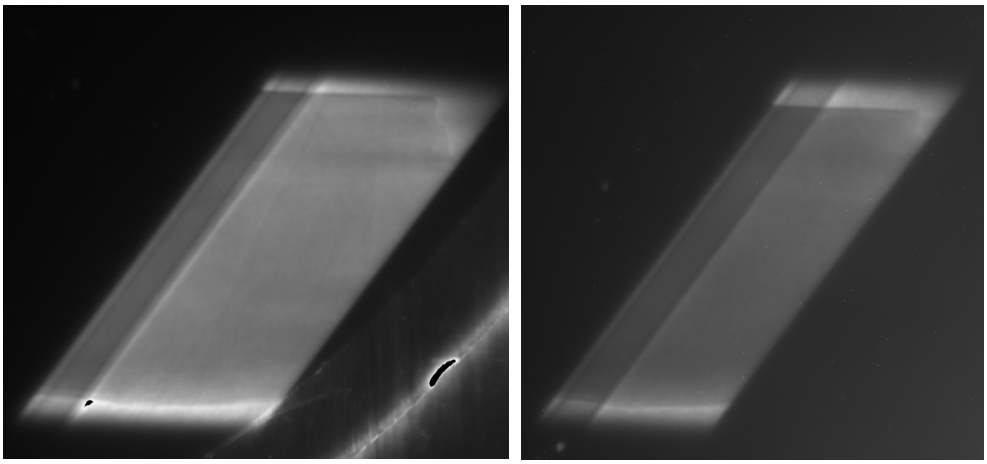

FIG. 1. X-ray images of Fe sample before and after the transition from bcc (left) to hcp (right) phase.

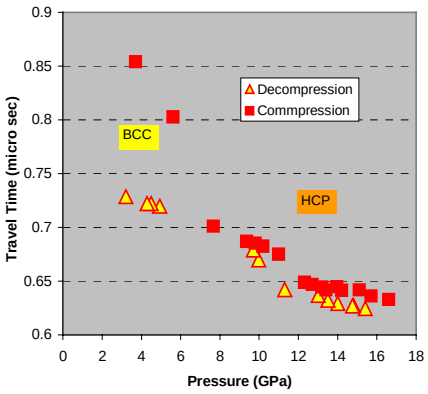

FIG. 2. S wave travel time as a function of pressure.

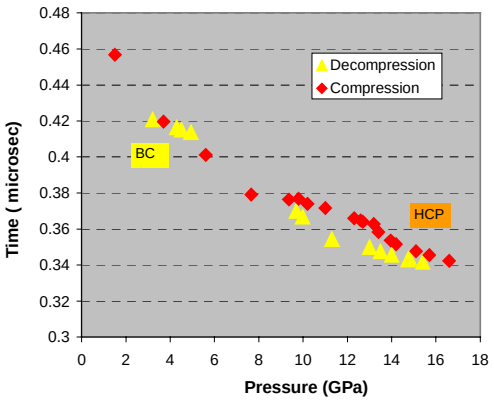

FIG. 3. P wave travel time as a function of pressure.

pressures) as well as a small degree of deformation along with cold compression. However, the strong attenuation of acoustic signals that was observed in this pressure range also suggests that this could have been a feature of the materials when the pressure approached the stability of the hcp phase. Between 8 and 10 GPa, the slow decrease of P and S wave travel times mostly reflected the effect of pressure on the metastable bcc phase. Travel-time discontinuities occurred in both the P and S waves at a pressure range of 10-14 GPa, marking the transition from the bcc to the hcp phase. It appears that the travel-time data had precursors before the phase transition detected by x-ray diffraction data.

The data collected during decompression were less affected by nonhydrostatic stresses after the sample was heated to a moderate temperature at each pressure. The P and S wave travel times showed pressure dependence similar to that of those in compression. The transition from the hcp to the bcc phase occurred at pressures of ~10.5 GPa, marked by a slope change in travel times. Compared to compression data, the phase transition had a hysteresis of about 2 GPa. X-ray diffraction data collected at pressures of ~10.2 GPa also confirmed the presence of the bcc phase. The velocity changes across the bcc-hcp phase transition at 10 GPa were 3% and 4%, respectively, for P and S waves. The velocities obtained from the current experiment are compatible with previously reported ultrasonic data [4] at 17 Gpa, within mutual uncertainties.

Acknowledgments

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