Powder x-ray diffraction of BaFe$_2$As$_2$ under hydrostatic pressure

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Abstract. We have performed powder x-ray diffraction study on BaFe$_2$As$_2$ under hydrostatic pressures up to 6 GPa at room temperature. Hydrostatic pressure was produced by means of a diamond anvil cell with He gas as a pressure transmitting medium. Single crystalline sample of BaFe$_2$As$_2$ was ground in alumina mortar cooled with liquid nitrogen to produce a fine powder sample. The diffraction patterns show a tetragonal structure with a space group of $I4_1/mmm$ in the whole pressure range. Rietveld analysis on the experimental results shows that the lattice parameters are monotonically decreased with increasing pressure. The As-Fe-As bond angle does not reach the ideal tetrahedral value of 109.47°.

1. Introduction

Family of FeAs-based superconductors includes several types of crystal structures with FeAs-layer. So-called 1111 system with the tetragonal ZrCuSiAs-type structure, like LaFeAs(O,F) firstly discovered in the family[1], shows the highest superconducting transition temperature $T_c$ at 55 K in SmFeAsO$_{1-x}$F$_x$[2]. In the 122 system with the tetragonal ThCr$_2$Si$_2$-type structure, potassium-doped Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$ exhibits the highest superconducting transition temperature, $T_c = 38$ K[3]. In both the 1111-system and the 122-system, parent compound without doping does not show superconductivity at ambient pressure. However, applying high pressure induces the superconductivity in both systems; LaFeAsO shows $T_c = 21$ K at 12 GPa[4] and BaFe$_2$As$_2$ shows $T_c = 35$ K at 3 GPa[5].

Non-doped BaFe$_2$As$_2$ does not exhibit superconductivity, but does have a spin density wave (SDW) transition at 140 K accompanied by a structural transformation from tetragonal ($I4/mmm$) at higher temperature to an orthorhombic ($Fmmm$) structure at lower temperature[6]. For the BaFe$_2$As$_2$ system, substituting of potassium for barium suppresses the SDW and structural transitions, and induces superconductivity at low temperatures. For non-doped BaFe$_2$As$_2$, application of pressure also suppresses both transitions, and superconductivity appears at low temperatures. Recently, we reported the pressure-induced superconductivity with $T_c = 35$ K at 3 GPa using a modified-Bridgman anvil-cell with Fluorinert as a pressure-transmitting medium (PTM)[5]. In other hand, pressurization using a cubic anvil cell with glycerin as a PTM, however, does not induce superconductivity up to 8 GPa[7]. More recently,
Yamazaki et al. reported that further increase in pressure gives the maximum $T_c$ value of 17 K at 11 GPa, using a cubic anvil cell with Daphne oil 7474 as a PTM[8]. Thus, appearance of the superconductivity of BaFe$_2$As$_2$ is strongly depends on the characteristics of the pressure apparatus and PTMs. In CaFe$_2$As$_2$, which has a same crystal structure as BaFe$_2$As$_2$, it is also reported that hydrostatic pressure produced by He gas as the PTM does not induce superconductivity up to 0.65 GPa[9], while the pressurization by a piston cylinder type cell with Fluorinert gives rise to superconductivity with $T_c \sim 10$ K around 0.5 GPa[10]. These results indicate that the appearance of the superconductivity in the 122 system, has close relationship with crystal structure and distortion caused by pressure.

In this paper, we report the results of the powder x-ray diffraction measurement under hydrostatic pressure using a diamond anvil cell (DAC) with He gas, up to 6 GPa to clarify structural change of BaFe$_2$As$_2$ under high pressure.

2. Experimental

Samples were prepared by the high-pressure synthesis method using a cubic-anvil-type apparatus (TRY-00120, Try Engineering Co. Ltd). Pure Ba (> 99%) and FeAs (99.5%) starting materials were mixed in appropriate amounts to prepare the nominal BaFe$_2$As$_2$ composition. Synthesis of BaFe$_2$As$_2$ was achieved by heating the mixture in a BN crucible under a pressure of approximately 3 GPa at 1473 K for 2 h. We could obtain a single-phase crystalline-sample, which were confirmed by observing a (00l) reflections from BaFe$_2$As$_2$ through x-ray diffraction using a Cu Kα line. The single crystalline sample put in an alumina mortar, was cooled in liquid nitrogen and ground into a fine powder.

The powdered sample was pressurized using a DAC assembled by a pair of type-Ia natural-diamond anvils with 0.6 mm culet diameters and 2 mm anvil heights. A rhenium foil was used as the gasket to make a sample chamber. The thickness of gasket was reduced from 120 to 60 μm by indentation with the diamond anvils. The gasket hole with a 260 μm diameter was made in the center of dent using an electron-discharge machine. The powdered sample was put in the chamber with ruby balls as a pressure marker. The chamber was filled with high-density He-gas, which was compressed up to 180 MPa at room temperature by a gas loading system of NIMS[11].

X-ray diffraction images were measured using synchrotron radiation light of Photon Factory in High Energy Accelerator Research Organization (KEK). Angle-dispersive diffraction-system in the beamline 18C was used for the measurement under high pressure. The sample in the DAC was irradiated using a highly monochromatized 20 keV synchrotron x-ray beam: the wavelength = 0.6191 Å. The beam diameter was 100 μm to avoid any contact of the x-ray beam with the gasket. Each diffraction image was recorded using a Fujifilm imaging plate with a resolution of 100 × 100 μm$^2$ and a size of 2000 × 2500 pixels. The exposure time was 30 min for each measurement. The pressure of sample was determined by the ruby fluorescence method[12]. The sample was pressurized up to 5.7 GPa.

3. Results and discussion

The x-ray diffraction measurements were carried out at several pressures up to 6 GPa at room temperature. Some of patterns are shown in Fig. 1. These patterns show that BaFe$_2$As$_2$ belongs to the space group of $I4/mmm$ in the whole pressure range. A diffraction peak shown beside the (101) peak, originates from unidentified impurity phase. We analyzed the obtained x-ray diffraction patterns using the RIETAN-2000 program[13], in which the diffraction patterns were fitted using the tetragonal $I4/mmm$ symmetry model. It is noted that a range around the peak of impurity phase is excluded from the fitting. Pressure dependence of the lattice parameter, $a$ and $c$, derived from the Rietvelt analysis, are shown in Fig. 2. This pressure dependence of the lattice parameters is almost same with the previous reports[14, 15].
Figure 1. X-ray diffraction patterns of BaFe$_2$As$_2$ under high pressure up to 6 GPa. Inverted triangles show the peak of impurity phase.

Figure 2. Pressure dependence of normalized lattice parameters for BaFe$_2$As$_2$.

Figure 3. Pressure dependence of relative unit-cell volume for BaFe$_2$As$_2$. The solid curve is a fit with the Murnaghan-type equation of state.

Figure 4. Pressure dependence of the As-Fe-As bond angle for BaFe$_2$As$_2$. z coordination of As atom is shown in upper panel.

Figure 3 shows the pressure dependence of the relative unit-cell volume for BaFe$_2$As$_2$. By fitting the pressure dependence of the relative volume with a Murnaghan-type equation of state, the values of the bulk modulus ($B_0$) and its pressure derivative ($B'_0$) are determined to be $65.4 \pm 1.4$ GPa and $4.4 \pm 0.6$, respectively. The pressure dependence of the volume up to 6 GPa does not show the phase transition to the pressure-induced volume-collapsed tetragonal phase, which appears above 0.35 GPa at 50 K for CaFe$_2$As$_2$[16] and above 27 GPa at 300 K for BaFe$_2$As$_2$[15], respectively. The As-Fe-As bond angle, which is of particular interest
for optimization of superconducting transition temperature[17], is shown in the bottom panel of Fig. 4. The two As-Fe-As angles are close to the ideal tetrahedral value, but does not reach 109.47° up to 6 GPa. Present results of the As-Fe-As angle under high pressure are not consistent with the previous reports by other groups[14, 15]. In their reports, the As-Fe-As angle shows very close value to the ideal tetrahedral angle around 3 GPa. In the top panel of Fig. 4, z coordination of As atom refined from the Rietvelt analysis was plotted as a function of pressure. The z coordination increase with increasing pressure. In Figs. 2, 3 and 4, another series of experimental results up to 4.8 GPa also shown as open symbols. These results agree well with the result shown as closed symbols. The secondary measurements were also performed in the same way as mentioned above; except for Inconel gasket (330 μm in diameter, 150 μm in thickness) and synchrotron wavelength (0.6196 Å).

In summary, we performed high-pressure powder x-ray diffraction studies for BaFe$_2$As$_2$ using the DAC with He gas as hydrostatic PTM up to 6 GPa. Refined results by Rietvelt analysis are summarized in Table 1 at selected pressures. The lattice parameters of BaFe$_2$As$_2$ are decreased almost monotonically with increasing pressure. The As-Fe-As angles are close to the ideal tetrahedral value, but does not reach 109.47° up to 6 GPa.

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Table 1. Refined results of the crystal structure for BaFe$_2$As$_2$ at selected pressures at room temperature. A parameter z shows atomic coordination of As atom in the space group I4/mmm: Ba (2a) (0 0 0), Fe (4d) (1/2 0 1/4), and As (4e) (0 0 z).

| Pressure | a[Å]     | c[Å]     | V[Å$^3$]  | z         | Rwp[%]   | S     |
|----------|----------|----------|-----------|-----------|----------|-------|
| 1 atm    | 3.9648(5)| 13.018(2)| 204.64(4) | 0.3535(3) | 2.37     | 1.90  |
| 0.4(1) GPa | 3.9576(5)| 12.981(2)| 203.31(4) | 0.3532(3) | 2.57     | 2.52  |
| 1.7(0) GPa | 3.9370(5)| 12.887(2)| 199.74(4) | 0.3539(3) | 3.36     | 3.20  |
| 3.5(1) GPa | 3.9098(5)| 12.751(2)| 194.92(4) | 0.3559(3) | 3.13     | 2.95  |
| 5.7(1) GPa | 3.8819(6)| 12.609(2)| 190.00(5) | 0.3560(2) | 2.93     | 2.49  |

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