PHYSICAL REVIEW B **79**, 172506 (2009)

Zero-resistance superconducting phase in $BaFe_2As_2$ under high pressure

Fumihiro Ishikawa,[1,*] Naoya Eguchi,[2] Michihiro Kodama,[2] Koji Fujimaki,[1] Mari Einaga,[1] Ayako Ohmura,[3]
Atsuko Nakayama,[3] Akihiro Mitsuda,[4] and Yuh Yamada[2]
[1]*Graduate School of Science and Technology, Niigata University, Niigata 950-2181, Japan*
[2]*Department of Physics, Niigata University, Niigata 950-2181, Japan*
[3]*Center for Transdisciplinary Research, Niigata University, Niigata 950-2181, Japan*
[4]*Department of Physics, Kyushu University, Fukuoka 812-8581, Japan*
(Received 21 February 2009; revised manuscript received 14 April 2009; published 27 May 2009)

A single-crystalline sample of $BaFe_2As_2$ was prepared by high-pressure synthesis, and temperature dependence of resistivity was measured under high pressures of up to 9.0 GPa. Application of 3.0 GPa pressure suppresses the spin-density wave transition and induces superconductivity with the zero resistance state at a superconducting transition temperature $T_c = 35$ K. T_c decreases monotonically with increasing pressure. The appearance of the superconductivity and the disappearance of the anomaly due to the spin-density wave transition occur simultaneously at a pressure of 3.0 GPa. At 3.0 GPa, a superconducting transition is induced in $BaFe_2As_2$; however, the superconductivity is suppressed at higher pressure.

DOI: 10.1103/PhysRevB.79.172506 PACS number(s): 74.25.Fy, 74.62.Fj, 74.70.Dd

Since the discovery of superconductivity in $LaFeAsO_{1-x}F_x$ at 26 K,[1] various superconductors with an iron-arsenide layer have been reported. Potassium-doped $Ba_{0.55}K_{0.45}Fe_2As_2$ with a $ThCr_2Si_2$-type structure, which also has an iron-arsenide layer, exhibits a superconducting transition at 38 K.[2] However, nondoped $BaFe_2As_2$ 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.[3,4]

For the $BaFe_2As_2$ system, the substitution of K for Ba suppresses the SDW and structural transitions and induces superconductivity at low temperatures.[2,3] For nondoped $BaFe_2As_2$, application of pressure also suppresses both transitions, and superconductivity appears at low temperatures. The Meissner effect is observed in $BaFe_2As_2$ under pressure by measurements of dc magnetization between 2.5 and 6.0 GPa.[5] The pressure dependence of the superconducting transition temperature T_c has a maximum value of 29 K near 4 GPa. The resistivity of $BaFe_2As_2$ drops below 20–30 K under high pressures above 3 GPa.[6] However, no zero resistance appeared under pressures of up to 13 GPa, while zero resistivity under high pressure was reported by various groups for the AFe_2As_2 (A=Ca, Sr) systems.[7–10]

Zero resistivity observations are important to clarify the intrinsic properties of pressure-induced superconductivity in the $BaFe_2As_2$ system. In this Brief Report, we present recent results of high-pressure synthesis and observation of zero resistance for the pressure-induced superconductivity in $BaFe_2As_2$.

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 $BaFe_2As_2$ composition. Synthesis of $BaFe_2As_2$ was achieved by heating the mixture in a BN crucible under a pressure of approximately 3 GPa at 1473 K for 2 h. A scanning electron microscope (SEM) image of the resulting sample is shown in Fig. 1. Powder x-ray diffraction (XRD) patterns measured using Cu $K\alpha$ radiation confirmed that the sample is single phase with the (001) orientation on the surface, which indicates a single-crystalline sample.

Pressures of up to 9.0 GPa were produced using a modified Bridgman anvil cell with a Teflon capsule.[11,12] The Teflon capsule has a sample space with an inner diameter of 1.5 mm. A mixture of Fluorinert FC-70 and 77 was used as the pressure-transmitting medium. Pressure was estimated from the load-pressure calibration curve measured in advance.[12] The calibration curve was obtained from the resistive transition of Bi due to the *I-II*, *II-III*, and *III-V* transitions at room temperature and the superconductive transition temperature of Pb measured by ac susceptibility at low temperature. Difference in the load-pressure curves between room and low temperatures is less than 0.1 GPa at same load.

The modified Bridgman-anvil-type cell was cooled using a liquid He-free-type 4 K cryocooler (Iwatani Industrial Gases Corp.). The electrical resistance under high pressures was measured by a conventional four-probe method. As electrical leads, gold wires, 20 μm in diameter, were used with a silver-loaded epoxy resin (Eccobond 56c, Emerson and Cuming) on the specimen.

The temperature dependence of resistivity in $BaFe_2As_2$ under high pressures up to 9.0 GPa is shown in Fig. 2. The

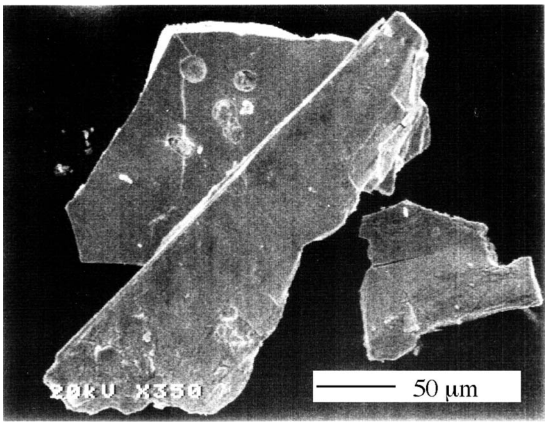

FIG. 1. SEM image of the $BaFe_2As_2$ sample.

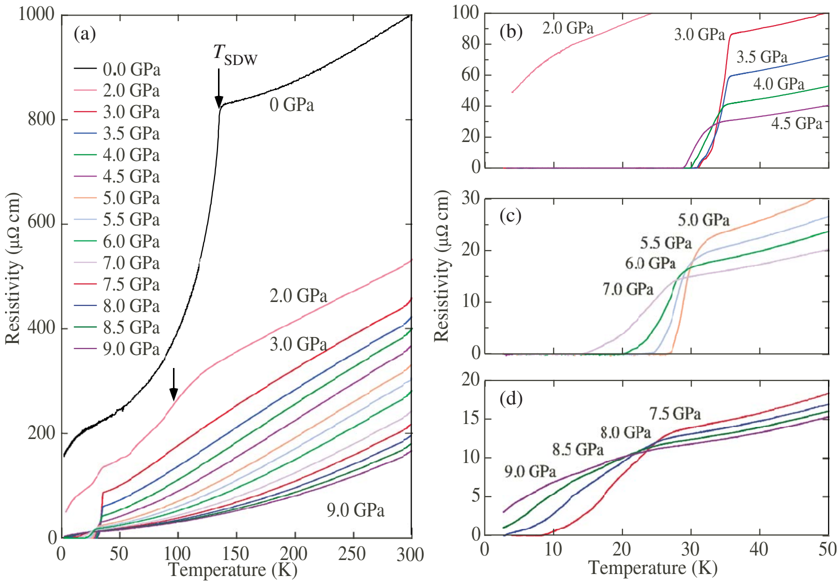

FIG. 2. (Color online) Temperature T dependence of the resistivity ρ in $BaFe_2As_2$ under pressure P; (a) below 300 K and at $0 \leq P \leq 9.0$ GPa, and below 50 K at (b) $2.0 \leq P \leq 4.5$ GPa, (c) $5.0 \leq P \leq 7.0$ GPa, and (d) $7.5 \leq P \leq 9.0$ GPa. Arrows indicate the SDW transition temperature derived from the peak of the temperature derivative of resistivity, $d\rho/dT$.

superconducting transition, with zero resistance, appears at 3.0 GPa, as shown in Figs. 2(a) and 2(b). At ambient pressure, an anomaly due to the SDW transition[3] appears at around $T_{SDW}=134$ K, which is derived from the peak of the temperature derivative of resistivity, $d\rho/dT$ (see the inset in Fig. 3). This temperature agrees well with the value of 131 K reported by Fukazawa *et al.*[6] At increased pressure of 2.0 GPa, the anomaly becomes broader and T_{SDW} decreases to 96 K. At 3.0 GPa, the superconducting transition with zero resistance state occurs at $T_c=35$ K, which is defined as the temperature of the onset of superconductivity. The superconducting transition is very sharp between 3.0 and 5.0 GPa, as shown in Figs. 2(b) and 2(c); the difference between T_c and the temperature where zero resistance state appears $T_{c-\text{zero}}$, is less than 5 K at 3.0 GPa. With increasing pressure, the transition temperature decreases monotonically, and the difference between T_c and $T_{c-\text{zero}}$ increases. At pressures above 7.5 GPa, no clear transition is observed and the zero resistance state disappears, as shown in Fig. 2(d). In more increased pressures of up to 9.0 GPa, $T_{c-\text{zero}}$ is not observed even at the lowest temperature in the present study, which is about 3 K. These results suggest that a pressure of 3.0 GPa induces the superconducting transition in $BaFe_2As_2$, whereas higher pressures simply suppress superconductivity. It is noted that the appearance of the superconductivity and the disappearance of the anomaly due to the SDW transition occur simultaneously at 3.0 GPa, as shown in Fig. 2(a). At 2.0 GPa, the SDW anomaly at 96 K and the resistivity drop at 35 K are observed; however, no zero resistance state appears, even at 3 K. Thus, the suppression of the SDW state is necessary for the appearance of the superconducting state with zero resistance in $BaFe_2As_2$.

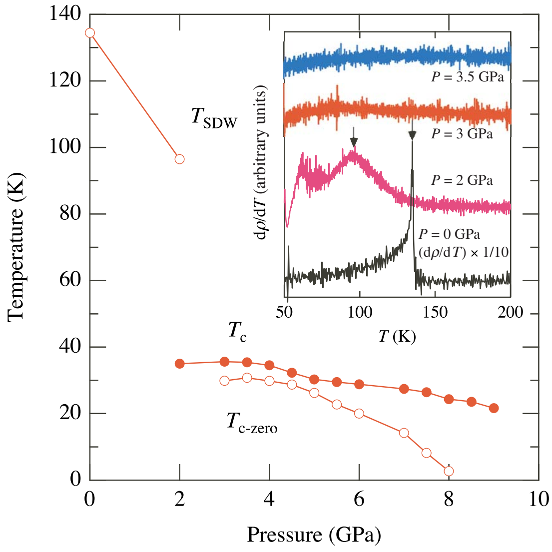

FIG. 3. (Color online) T-P phase diagram of $BaFe_2As_2$ determined from resistivity anomalies under high pressure. Inset shows $d\rho/dT$ vs T in $BaFe_2As_2$ under high pressure. Arrows indicate the SDW anomaly derived from the peak of $d\rho/dT$.

Magnetization measurements of $BaFe_2As_2$ under high pressures indicate that the superconducting transition occurs gradually with increasing pressure, while the superconducting transition in $SrFe_2As_2$ occurs abruptly, accompanied by a maximum in superconducting temperature.[5] The present results for the pressure dependence of T_c for $BaFe_2As_2$ are similar to those for $SrFe_2As_2$ (Ref. 5) and $CaFe_2As_2$ (Ref. 8) in terms of the appearance of superconductivity; the onset of superconductivity occurs abruptly at a critical pressure.

The pressure dependences of T_c, $T_{c-\text{zero}}$, and T_{SDW} are presented in Fig. 3. T_c and $T_{c-\text{zero}}$ decrease with increasing pressure, and both the SDW anomaly and superconductivity are suppressed by pressure. The pressure dependence of T_c shown in Fig. 3 is very similar to that for the characteristic temperature of polycrystalline $BaFe_2As_2$.[6] However, for the polycrystalline sample, the resistivity drops sharply at the characteristic temperature, which probably corresponds to T_c, at the pressure of 13.0 GPa; the difference between T_c and $T_{c-\text{zero}}$ is small even in the pressures above 9 GPa. Moreover, the pressure dependence of T_c obtained from the magnetization measurements up to the pressure of 6 GPa (Ref. 5) shows little change in T_c between 5 and 6 GPa. Thus, T_c and $T_{c-\text{zero}}$ behavior shown in Fig. 3 is not consistent with the previous reports,[5,6] especially in high-pressure region above 5 GPa.

Several authors reported the effect of pressure on the superconductivity of the carrier-doped $BaFe_{2-x}Co_xAs_2$ and $Ba_{1-x}K_xFe_2As_2$ systems.[13,14] Resistivity measurements of $BaFe_{1.92}Co_{0.08}As_2$ under high pressures show that the SDW transition is not completely suppressed up to 2.5 GPa and T_c increases slightly with increasing pressure.[13] Moreover, T_c is almost independent of pressure in $BaFe_{1.8}Co_{0.2}As_2$, which shows no anomaly due to the SDW transition.[13] In contrast, an opposite effect of pressure on T_c was reported for the $Ba_{1-x}K_xFe_2As_2$ system; T_c decreases with increasing pressure.[14] Resistivity measurements of $Ba_{1-x}K_xFe_2As_2$ showed that the SDW state is suppressed by K doping. Thus, in the $BaFe_2As_2$ and related systems, T_c decreases with increasing pressure when the SDW transition is completely suppressed by pressure or doping. Consequently, T_c on these systems under high pressure depends on the competition between two kinds of pressure effects on superconductivity: direct suppression effect and enhancement effect via suppression of the SDW state. These results may support such a scenario as follows: (1) the superconductivity of $BaFe_2As_2$ and related systems is suppressed by the SDW state. (2) The suppression of the SDW state by application of pressure results in onset of the superconductivity. (3) Further increase in pressure suppresses the superconductivity after the SDW state is completely suppressed.

In summary, a single-crystalline sample of $BaFe_2As_2$ was prepared by high-pressure synthesis using a cubic-anvil-type apparatus, and the temperature dependence of resistivity was measured under high pressures up to 9.0 GPa. Application of 3.0 GPa pressure suppresses the SDW transition and induces superconductivity with the zero resistance state at $T_c = 35$ K. T_c decreases monotonically with increasing pressure, and no clear transition is observed above 7.5 GPa. The appearance of the superconductivity and the disappearance of the SDW transition anomaly occur simultaneously at a pressure of 3.0 GPa. The effect of pressure suppresses both the SDW state and superconductivity in $BaFe_2As_2$.

The authors wish to thank T. C. Ozawa and Y. Ōno for valuable discussions and advice. This research was partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture of Japan, and a Grant for Promotion of the Niigata University Research Projects.

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*fumihiro@phys.sc.niigata-u.ac.jp

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