

# Appearance of pressure-induced superconductivity in $\text{BaFe}_2\text{As}_2$ under hydrostatic conditions and its extremely high sensitivity to uniaxial stress

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We have performed electrical resistivity measurements on single-crystal  $\text{BaFe}_2\text{As}_2$  under high pressure  $P$  up to 16 GPa with a cubic anvil apparatus, and up to 3 GPa with a modified Bridgman anvil cell. The samples were obtained from the same batch, which was grown with a self-flux method. A cubic anvil apparatus provides highly hydrostatic pressure, and a modified Bridgman anvil cell, which contains liquid-pressure transmitting medium, provides quasihydrostatic pressure. For highly hydrostatic pressure, the crystal phase and magnetic-transition temperature decreases robustly with  $P$  and disappears at around 10 GPa. The superconducting phase appears adjacent to magnetic phase in narrow pressure region between 11 and 14 GPa. The tiny difference in hydrostaticity between the cubic anvil apparatus and modified Bridgman anvil cell induces a drastic effect on the phase diagram of  $\text{BaFe}_2\text{As}_2$ . This result indicates that small uniaxial stress along  $c$  axis strongly suppresses the structural/antiferromagnetic ordering and stabilizes superconductivity at much lower pressure.

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## I. INTRODUCTION

The discovery of superconductivity in F-doped  $\text{LaFeAsO}$  at 26 K by Kamihara *et al.*<sup>1</sup> heralded a new era of superconductivity based on iron. The common feature of the new superconductors is existence of iron pnictide layers, which is analogous to  $\text{CuO}_2$  planes of the cuprate superconductors. After extensive investigations, high-temperature superconductivity with a transition temperature  $T_c$  of 56 K was observed in rare-earth iron oxypnictides (1-1-1-1 compound).<sup>2,3</sup> Members of superconducting iron family were observed one after another in oxygen free compounds such as  $\text{BaFe}_2\text{As}_2$  having  $\text{ThCr}_2\text{As}_2$ -type crystal structure (1-2-2 compound),<sup>4,5</sup>  $\text{LiFeAs}$  (1-1-1 compound),<sup>6</sup> and  $\text{FeSe}$  (1-1 compound).<sup>7</sup> New compounds having a thick perovskite layer were also reported recently.<sup>8</sup> Among them, the 1-2-2 compound  $A\text{Fe}_2\text{As}_2$  ( $A=\text{Ba}, \text{Ca}, \text{and Sr}$ ) occupies a singular position, which is particularly suitable for precise physical property measurements, since high-quality large single crystals can be grown in the congruent melting condition. The 1-2-2 compounds have a first-order antiferromagnetic (AF) transition on cooling at moderately high temperature, which always accompanied by a phase transition from tetragonal to orthorhombic structure. In the case of  $\text{BaFe}_2\text{As}_2$ , the crystal structure/magnetic phase transition occurs at 141 K.<sup>9</sup> The carrier doping of K for Ba, or Co for Fe strongly suppresses the transition in  $\text{BaFe}_2\text{As}_2$ . The highest  $T_c$  of 38 K in the 1-2-2 compounds is obtained in a K-doped material located adjacent to AF state.<sup>5</sup>

Another method to suppress AF ordering is applying pressure  $P$ . This has a great advantage over chemical doping since the ground state of the system can be tuned without introducing any disorder. Hence, many high-pressure measurements have been performed in iron pnictides.<sup>10-20</sup> However, contradicting results were reported in the 1-2-2 compound up to now. The simplest reason is sample quality

difference. Polycrystalline samples are inhomogeneous and have broader phase transitions than single crystals. Even in single crystals, the sample growth condition makes a difference, i.e., the crystals grew up in Sn or self-flux. The contamination by other elements from the flux often influences the physical properties. Most reliable results are obtained in single crystals made by self-flux method. Even though high-quality crystals were used, many groups have reported scattered and confusing results under pressure. The discrepancy may be induced by the difference in the hydrostaticity in respective measurements.

The effect of pressure inhomogeneity has been studied in  $\text{CaFe}_2\text{As}_2$  (Ref. 19) and  $\text{SrFe}_2\text{As}_2$ .<sup>13</sup> Duncan *et al.*<sup>20</sup> have studied the difference in hydrostaticity in  $\text{BaFe}_2\text{As}_2$  using a piston cylinder cell, an alumina anvil cell with Daphne oil and a Bridgman anvil cell with solid steatite as pressure-transmitting medium. They reported that uniaxial stress strongly suppresses structural/AF ordering and induces superconductivity at lower pressure. Even if the importance of uniaxial stress was pointed out by Duncan *et al.*, phase diagram of pressure-induced superconductivity in hydrostatic condition was not reported. Hence, we have studied the difference in hydrostaticity in highly homogeneous region to settle the confusion of the pressure induced phenomena in  $\text{BaFe}_2\text{As}_2$ .<sup>10,11,14-16,20</sup> We performed resistivity measurements using self-flux grown single crystals of  $\text{BaFe}_2\text{As}_2$  from the same batch up to 16 GPa with a cubic anvil apparatus and compared the results with those obtained with a Bridgman anvil cell containing liquid-pressure transmitting medium.

## II. EXPERIMENTAL

Single crystals of  $\text{BaFe}_2\text{As}_2$  were grown by the FeAs self-flux method to avoid contamination. The starting materials, Ba and FeAs, were put into an alumina crucible with a ratio

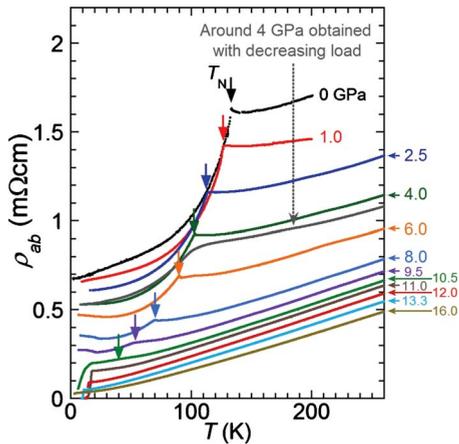


FIG. 1. (Color online)  $T$  dependence of electrical resistivity in  $\text{BaFe}_2\text{As}_2$  single crystal under various pressures obtained by a cubic anvil apparatus. In addition to the above data, resistivity around 4 GPa, which was obtained in decreasing load process, was also plotted.

of 1:4 and sealed in evacuated quartz tube. The tube was heated up to 1140 °C and cooled down very slowly to 1040 °C. Details of the sample preparation have been previously reported.<sup>21</sup> X-ray analysis showed that the crystals had a tetragonal  $\text{ThCr}_2\text{Si}_2$ -type structure with no impurity phase. The cubic anvil high-pressure apparatus consists of six anvils and homogeneously compresses the gasket cube containing a Teflon capsule.<sup>22</sup> The sample is located inside the Teflon cell together with liquid-pressure-transmitting medium Daphne oil 7474.<sup>23</sup> The cubic anvil apparatus has an ability to make a highly hydrostatic pressure even if liquid-pressure-transmitting medium becomes solid at high-pressure region as cubic-shaped gasket is compressed by six anvils in all cubic faces. The pressure of the sample is calibrated by the resistivity anomalies of Bi, Te, Sn, and ZnS associated with their structural phase transitions at room temperature. The weight loaded to the sample is kept constant during the same pressure measurement. This apparatus produces a very homogeneous and hydrostatic pressure up to 20 GPa. In the modified Bridgman cell,<sup>24</sup> the sample is in a Teflon cell together with a liquid-pressure-transmitting medium, a 1:1 mixture of Fluorinerts 70 and 77. It is expected that the solidification of Fluorinerts above 1.2 GPa yields moderate inhomogeneous pressure distributions. The modified Bridgman cell provides less hydrostatic condition than the cubic anvil apparatus. The pressure of the sample in Bridgman cell is also calibrated by the resistivity anomalies of Bi and Te at room temperature, and additionally from the  $T_c$  of Pb at low temperatures. The electrical resistivity measurements under pressure were performed by a standard dc four-probe technique with a current flow in the  $ab$  plane. The electric leads were gold wire of 20  $\mu\text{m}$  in diameter with a silver-loaded epoxy resin to contact the crystal.

### III. RESULTS AND DISCUSSION

Figure 1 shows the temperature  $T$  dependence of the electrical resistivity  $\rho(T)$  of  $\text{BaFe}_2\text{As}_2$  under pressure obtained

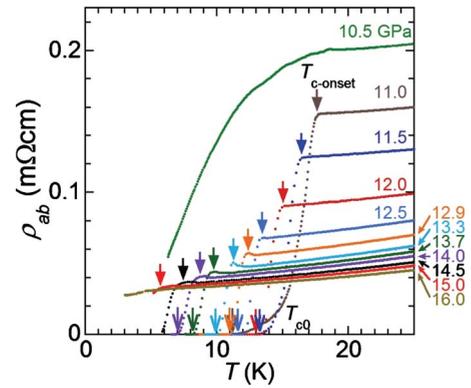


FIG. 2. (Color online) Expanded view of resistivity above 10 GPa at low temperature. The onset of  $T_c$ ,  $T_{c\text{-onset}}$ , is defined by anomaly in  $d\rho/dT$ , and  $T_{c0}$  is defined by temperature at which  $\rho$  becomes zero.

using the cubic anvil apparatus. At ambient pressure,  $\rho(T)$  decreases gradually on cooling from 300 K, and decreases steeply just below 134 K as reported by many groups. The anomaly in the resistivity, determined by the maximum of  $d\rho/dT$ , corresponds to the crystal structure and AF transition. We determined the transition temperature  $T_N$  from this anomaly.<sup>25</sup>  $T_N$  decreases with a slope of  $dT_N/dP = -7.0$  K/GPa. The change in the transition is robust against pressure, diminishing gradually and disappearing above 10 GPa. Our present result below 8 GPa is similar to that obtained in a single crystal with a cubic anvil apparatus by Matsubayashi *et al.*<sup>14</sup> and is also consistent with that previously obtained in polycrystalline samples.<sup>11</sup> An expanded view of resistivity above 10 GPa at low temperatures is shown in Fig. 2.  $\rho(T)$  rapidly decreases below approximately 17 K and becomes zero at lower temperatures at 11 GPa, which indicates the collapse of structural/AF ordering and appearance of superconductivity. A drop of the resistivity to zero appears at 11 GPa. The transition becomes sharp at 11.5 GPa. The  $T_{c0}$ , defined by temperature at which  $\rho$  becomes zero, of 13 K is the highest at 11.5 GPa, decreases monotonically with increasing  $P$ , and zero resistivity was not achieved down to 3 K at 16 GPa.

The electrical resistivity measurement itself is insufficient and further susceptibility or NMR measurements are needed to examine the bulk nature of the superconductivity. In  $\text{SrFe}_2\text{As}_2$ , ac susceptibility measurement in a cubic anvil apparatus by Matsubayashi *et al.*<sup>14</sup> has shown the appearance of bulk superconductivity in a very narrow region which is adjacent to AF phase. The bulk nature of superconductivity in  $\text{SrFe}_2\text{As}_2$  was also examined by  $^{75}\text{As}$  NMR under hydrostatic high-pressure condition, which was performed using a soft solid Ar as pressure transmitting medium by Kitagawa *et al.*<sup>26</sup> We expect that the situation of  $\text{BaFe}_2\text{As}_2$  would be similar to that of  $\text{SrFe}_2\text{As}_2$ . In the cubic anvil apparatus, very small inhomogeneity in pressure exists when six anvils are not identically pressed. The pressure dependence of transition width associated with  $T_c$  is one way to evaluate the pressure inhomogeneity. Using the same cubic anvil apparatus, the pressure dependence of  $T_c$  was measured in Fe superconductor  $\text{NdFeAsO}_{1-y}$  up to 18 GPa.<sup>27</sup> The transition

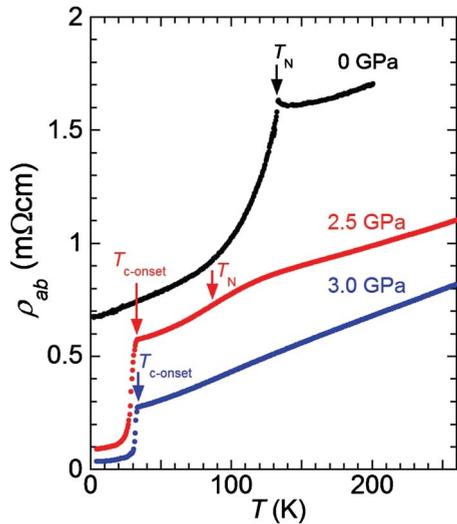


FIG. 3. (Color online)  $T$  dependence of resistivity obtained by a modified Bridgman anvil cell.

width associated with  $T_c$  remains narrow even at 18 GPa. Such narrow transitions in Fe superconductor were not reported previously. The cubic anvil apparatus is one method to acquire homogeneous pressure beyond 10 GPa.

In order to study the inhomogeneity of pressure, we measured  $\rho(T)$  of  $\text{BaFe}_2\text{As}_2$  from the same batch by a modified Bridgman anvil cell. The crystal was aligned with its  $c$  axis perpendicular to the anvil surfaces. As the sample was in the liquid-pressure-transmitting medium Fluorinerts, the uniaxial stress would be more or less reduced. However, solidification of Fluorinerts occurs above 1.2 GPa at room temperature, which reduces pressure homogeneity at higher pressure.

The result obtained by the modified Bridgman anvil cell is shown in Fig. 3. The anomaly at  $T_N$  is soon broadened under pressure.  $T_N$  decreases faster than that of the cubic anvil apparatus. Another lower  $T$  anomaly was observed at 30 K above 2.5 GPa. The rapid drop of electrical resistivity indicates appearance of superconductivity. However, nonzero resistivity below the transition shows that the induced superconductivity is not bulk but filamentary. This anomaly around 30 K was often observed by many groups.<sup>11,15,16</sup> Our results at low pressure agree with the  $T_{c\text{-onset}}$  data obtained by Ishikawa *et al.* using a modified Bridgman anvil cell. These results are summarized and plotted in Fig. 4. Usually, the small inhomogeneous pressure in a modified Bridgman cell has little or no effect. However, an inhomogeneous pressure gives rise to huge effect in the phase diagram of  $\text{BaFe}_2\text{As}_2$ , which is not accounted by the simple pressure inhomogeneity.

Let us consider why  $\text{BaFe}_2\text{As}_2$  is so sensitive to uniaxial stress. The x-ray measurement<sup>5,28</sup> shows that the lattice constants of  $\text{BaFe}_2\text{As}_2$  for the  $a$ ,  $b$ , and  $c$  axes ( $a, b, c$ ) have a characteristic feature. The  $T$  dependence of  $a$ ,  $b$ , and  $c$  are highly anisotropic and completely different in the tetragonal and the orthorhombic structures. In the tetragonal structure,  $c$  decreases and  $a(b)$  remains constant with decreasing  $T$ . On the contrary, in the orthorhombic structure the average of  $a$  and  $b$  decreases, and  $c$  becomes constant with decreasing  $T$  i.e., the volume reduction in the tetragonal structure occurs

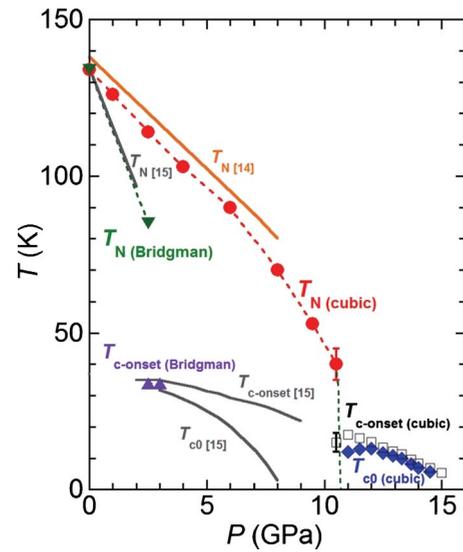


FIG. 4. (Color online)  $P$ - $T$  phase diagram of  $\text{BaFe}_2\text{As}_2$  obtained by a cubic anvil apparatus (cubic) and a modified Bridgman anvil cell (Bridgman). We also plot the  $T_N$  reported by Matsubayashi *et al.* (Ref. 14) using a cubic anvil apparatus and the  $T_N$ ,  $T_{c\text{-onset}}$ , and  $T_{c0}$  reported by Ishikawa *et al.* (Ref. 15) using a modified Bridgman anvil cell.

through the shrinkage along  $c$  axis, and that in orthorhombic structure through shrinkage along the  $a$  and  $b$  axes on cooling. A similar result for the  $T$  dependence of lattice parameters is observed in thermal expansion.<sup>29</sup> Then we consider the situation where uniaxial stress is applied to the crystal along the  $c$  axis, which is plotted schematically in Fig. 5. When uniaxial stress is applied, the free energy is minimized by the crystal shrinking along the  $c$  axis. The tetragonal structure is stabilized by uniaxial stress, which explains the high sensitivity to the inhomogeneity of pressure. The importance of uniaxial stress, which increases  $c/a$  ratio, in the structural transition from tetragonal to orthorhombic was pointed out by Duncan *et al.*<sup>20</sup> This effect is very serious even in quasi-hydrostatic pressure condition. The peculiar temperature dependences of lattice parameters in both structures clearly show stabilization of tetragonal phase under uniaxial condition. In this way, a small amount of “tetragonal structure phase induced by inhomogeneous pressure” develops as small islands in the sea of orthorhombic structure/AF phase in the lower-pressure region. Superconductivity appears in the induced tetragonal structure phase at  $T_c$  probably

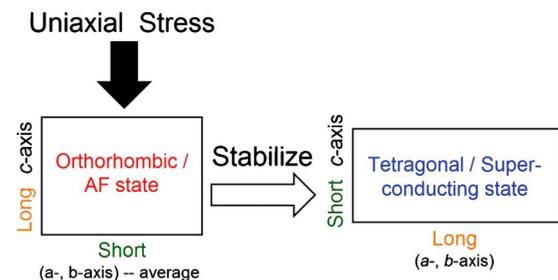


FIG. 5. (Color online) Schematic view of role of uniaxial stress in  $\text{BaFe}_2\text{As}_2$ .

determined from so-called “Lee plot” which is empirical relation between  $T_c$  and As-Fe-As bond angle.<sup>30</sup> As seen in Fig. 4, the  $T_{c \text{ onset}}$  is smooth having a maximum value at around 38 K at low pressure and finally seems to disappear around 16 GPa, which is similar to the  $P$  dependence of  $T_c$  observed in  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ .<sup>31</sup> Indeed, the tendency of the As-Fe-As bond angle moving away from the highest- $T_c$  angle with increasing  $P$  is already reported by Kimber *et al.*<sup>32</sup> In order to establish the  $P$ - $T$  phase diagram of  $\text{BaFe}_2\text{As}_2$ , more precise structural study under pressure in which pressure condition is the same as that utilized in resistivity measurement is desirable.

#### IV. CONCLUSION

In conclusion, we have measured the electrical resistivity of single-crystal  $\text{BaFe}_2\text{As}_2$  under high pressure with a cubic anvil apparatus and with a modified Bridgman anvil cell to determine the  $P$ - $T$  phase diagram. Under highly hydrostatic conditions, the crystal structure/AF transition temperature

decreases robustly against pressure and disappears around 10 GPa. Superconductivity appears in the pressure range between 11 and 14 GPa. The highest  $T_{c0}$  of 13 K is obtained at 11.5 GPa, which is probably adjacent to the orthorhombic/AF phase, and  $T_c$  decreases monotonically with increasing  $P$ . Uniaxial stress, which is induced by tiny departure from hydrostaticity, strongly suppresses structural/AF ordering, and stabilizes superconductivity.

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