Upper Critical Field of Pressure-Induced Superconductor EuFe$_2$As$_2$

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We have carried out high-field resistivity measurements up to 27 T in EuFe$_2$As$_2$ at $P = 2.5$ GPa, a virtually optimal pressure for the $P$-induced superconductivity, where $T_c = 30$ K. The $B_{c2} - T_c$ phase diagram has been constructed in a wide temperature range with a minimum temperature of 1.6 K ($\approx 0.05 \times T_c$), for both $B || ab$ ($B_{c2}^{ab}$) and $B || c$ ($B_{c2}^c$). The upper critical fields $B_{c2}^{ab}(0)$ and $B_{c2}^c(0)$, determined by the onset of resistive transitions, are 25 T and 22 T, respectively, which are significantly smaller than those of other Fe-based superconductors with similar values of $T_c$. The small $B_{c2}(0)$ values and the $B_{c2}(T)$ curves with positive curvature around 20 K can be explained by a multiple pair-breaking model that includes the exchange field due to the magnetic Eu$^{2+}$ moments. The anisotropy parameter, $\Gamma = B_{c2}^{ab}/B_{c2}^c$, in EuFe$_2$As$_2$ at low temperatures is comparable to that of other “122” Fe-based systems.

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The discovery of superconductivity in LaFeAs(O,F) at $T_c = 26$ K$^1$ has inspired experimental and theoretical research on a group of FeAs-layered superconductors (SCs)$^{2}$ Basically, Fe-based high-$T_c$ superconductivity$^{3}$ occurs when the antiferromagnetic (AF) order in the mother compounds is suppressed by means of carrier doping$^{4}$ or isovalent substitution$^{5}$. As compared to other methods in studying such interplay between magnetism and superconductivity, pressure experiments have a significant advantage in that they are free from random impurity potentials that may distort the underlying physics of the low-lying energy states. Among the various crystal structures, tetragonal ThCr$_2$Si$_2$-type (“122”) compounds have been investigated more intensively owing to the availability of highly-pure stoichiometric single crystals. In particular, AFe$_2$As$_2$ (A = Sr, Eu) exhibits $P$-induced bulk superconductivity$^{6,7}$ at $T_c$ of order 30 K$^8$. In contrast, superconductivity under hydrostatic pressure is not exhibited by CaFe$_2$As$_2$$^9$ and its occurrence in BaFe$_2$As$_2$ has not been established definitively.\textsuperscript{10,11}

A fundamental characteristic of SCs is the upper critical field $B_{c2}$. $B_{c2}$ has its roots in the breakdown of Cooper pairs; hence, the $B_{c2} - T_c$ phase diagram provides important insights into the pairing mechanism of high-$T_c$ superconductivity. Thus far, to our knowledge, there has been no reports on $B_{c2}$ for $P$-induced Fe-based SCs at low temperatures. This is mainly attributed to the difficulty in conducting high-pressure experiments on high-$T_c$ SCs under a high-field. In the case of SrFe$_2$As$_2$ ($T_c = 30$ K at 4.2 GPa), a field of 8 T brings about a small reduction in $T_c$ (i.e., to 27 K) for $B || ab$.$^{12}$ Assuming an orbitally limited case$^{13} B_{c2}(T = 0 \text{ K})$ could exceed 60 T.$^{14}$ However, the low-temperature region of the $B_{c2}$ curve, where paramagnetic and/or multiband effects may play important roles$^{15,16}$ has not been investigated.

In the case of EuFe$_2$As$_2$ ($T_c = 30$ K at ~2.5 GPa), $B_{c2}$ is relatively small, i.e., ~16 T between 5 K and 10 K$^2$. and hence can be traced down to very low temperatures. EuFe$_2$As$_2$ is unique in that the localized Eu$^{2+}$ moments exhibit an AF order below 20 K$^2$. In addition to an AF order arising from the FeAs layers at $T_0 \sim 190$ K, $T_N$ of the Eu$^{2+}$ moments is insensitive to pressure, and the AF order occurs in the $P$-induced superconducting state as evidenced by magnetic and heat capacity measurements under high-pressure.$^{9,18-20,23,24}$

Thus, EuFe$_2$As$_2$ provides an excellent opportunity where a long-standing issue of the interplay between superconductivity and magnetism can be studied in a high-$T_c$ material using high-quality single crystals.

In this report, we present the $B_{c2} - T_c$ phase diagram of EuFe$_2$As$_2$ at a pressure of 2.5 GPa and minimum temperature of 1.6 K via high-field resistivity measurements up to 27 T, and discuss the origin of the distinctive $B_{c2}$ curves.

Single crystals of EuFe$_2$As$_2$ were prepared via the Bridgman method from a stoichiometric mixture of the constituent elements. The samples analyzed in this study were obtained from the same batch (residual resistivity ratio $RRR = 7$) as that used in Refs.\textsuperscript{21} and \textsuperscript{22}. The resistivity of two samples, denoted by #1 and #2, was simultaneously measured at $P = 2.5$ GPa via an ac four-probe method in a $^4$He cryostat ($T \geq 1.6$ K). Sample #1 ( #2) was aligned with the $ab$-plane (c-axis) parallel to the longitudinal direction of a hybrid-type piston cylinder pressure cell$^{25}$ for $B || ab$ ($c$) measurements. For both
samples, the magnetic field was applied along the piston cylinder axis in a direction perpendicular to that of the current. To generate hydrostatic pressure, Daphne 7474 (Idemitsu Kosan) oil, which remains in the liquid state up to 3.7 GPa at room temperature, was used as the pressure-transmitting medium. The samples were gradually cooled at an average rate of 0.5 K/min. The pressure was calibrated at 4.2 K by the resistance change of a Manginian wire. Magnetic fields up to 27 T were produced by a water-cooled resistive magnet installed at the Tsukuba Magnet Laboratory, National Institute for Materials Science. A 17-T superconducting magnet was used for preliminary resistivity studies. In this study, the magnetic field \( B \) denotes an externally applied field, and the magnetization within a sample (up to \( \sim 0.9 \text{T} \)) is neglected.

Figure 1 shows the temperature dependence of the resistivity, \( \rho(T) \), for the two samples, #1 and #2, at \( P = 2.5 \text{ GPa} \) in the absence of an applied field. For both samples, \( \rho \) exhibits virtually \( T \)-linear dependence in the broad temperature range above \( T_c \) without any anomaly due to the AF order arising from the FeAs layers and localized \( \text{Eu}^{2+} \) moments. This observation is consistent with the phase diagram shown in the inset. For both samples, \( P = 2.5 \text{ GPa} \) is just above the critical pressure \( P_c \), where \( T_0 \rightarrow 0 \), as indicated by the arrow. Similar \( \rho \sim T \) behavior was also reported in several optimally doped Fe-based SCs. However, the reason for such behavior has not been verified thus far. Both samples exhibit a sharp transition to zero resistivity at \( T_c = 30 \text{ K} \); the reentrant-like behavior as reported in Ref. 20 is not observed for either sample at this pressure. Our previous work indicates that reentrant-like behavior may be observed for \( P < P_c \) but not for \( P > P_c \) (as long as \( P \) is not far from \( P_c \)) in our single crystals. Since both \( T_c \) and \( B_{c2} \) attain maximum values at \( P \approx P_c \), followed by a monotonic decrease with increasing \( P \), \( B_{c2} \) determined at 2.5 GPa in this study is expected to be close to its maximum value.

Figure 2(a)-(d) shows the resistivity of EuFe\(_2\)As\(_2\) at 2.5 GPa as a function of \( T \) and \( P \). A magnetic field of 27 T is sufficient to recover the normal state at the minimum temperature, 1.6 K (\( \approx 0.05 \times T_c \)), for both orientations. Using the data in Fig. 2(a) and (b), the \( B_{c2} - T_c \) phase diagram of EuFe\(_2\)As\(_2\) is constructed for \( B || ab \) at 2.5 GPa, as shown in Fig. 3. Three sets \(-B_{c2}, B_{c2}^\parallel (\text{onset}), \) and \( B_{c2}^\parallel (x = 0 \text{ and } 50, \text{ x\% of the normal state resistivity } \rho_n) \) are plotted, and their definitions are illustrated in the inset. The solid and open symbols are obtained from the \( \rho(B) \) and \( \rho(T) \) measurements, respectively. \( B_{c2}^\parallel \) consistent with the previous result \((\times)\) obtained from the ac-\( \chi \) measurement for \( B || ab \). Note that all the curves of \( B_{c2} \) for \( B || ab (B_{c2}^\parallel) \) obtained under different criteria exhibit qualitatively similar \( T \)-dependence. The same is also true for \( B_{c2} \) for \( B || c (B_{c2}^\parallel) \), as shown in Fig. 4(a). \( T_N \) at zero field is indicated by an arrow in Figs. 3 and 4. However, we note that, since the AF order of the \( \text{Eu}^{2+} \) moments is destroyed by an applied field of \( \sim 1 \text{T} \), the \( B_{c2} \) curves for both \( B || ab \) and \( B || c \) are in the paramagnetic or field-induced FM state of the \( \text{Eu}^{2+} \) moments.

A distinctive feature, the concave (upward) curvature of \( B_{c2} \) around 20 K, has not been reported in other Fe-based SCs without localized magnetic ions. Therefore, it is likely related to the magnetic state of the \( \text{Eu}^{2+} \) moments. Similar concave \( B_{c2}(T) \) curves have been reported in Chevrel-phase compounds such as \((\text{Eu},M)\text{Mo}_6\text{S}_8\) \((M = \text{Sn}, \text{La}, \text{etc.})\) and \(\text{EuMo}_6\text{S}_8\) under pressure. In these systems, the conduction electrons are subjected to an exchange field \( B_J \) in addition to an
applied field via AF coupling with the Eu$^{2+}$ localized magnetic moments. Note that the concave curvature is an indication of the negative sign of $B_J$; $B_J$ is antiparallel to the applied field. Within a multiple pair-breaking picture, $B_{c2}$ in the dirty limit of three-dimensional SCs is a consequence of a large Eu$^{2+}$ magnetic susceptibility as $T \to T_c$, which improves the fit in a way to phenomenologically overcome this problem and to describe the experimental curves excellently over the entire range of magnetic fluctuations may be detrimental to superconductivity. One might expect that the low value of $B_{c2}$ (compared to other Fe-based SCs’ with similar $T_c$ values) is due to the large $B_J$, which is a consequence of a large Eu$^{2+}$ magnetization due to the field-induced FM alignment of the Eu$^{2+}$ moments. However, its deviation from the experimental curve is also noticeable at low fields near $T_c$. This disagreement probably indicates that the phase diagram in this $T$-range is affected by a subtle competition between superconductivity and magnetic fluctuations, and it is beyond the scope of Eq. (1), which assumes a homogeneous $B_J$ produced by paramagnetic spins. Since the dominant interaction among the Eu$^{2+}$ moments is the intralayer FM interaction, the FM fluctuations develop when $T \to T_N$, and this is shown by the enhanced of the magnetic susceptibility as $T \to T_N$ in EuFe$_2$As$_2$, determined by the onset and zero-resistivity. The dashed curves are fits for $T = 0$ extrapolation (see text). (b) $T$-variation of an anisotropy parameter, $\Gamma = B_{c2}^\|/B_{c2}^\perp$, determined by the onset and zero-resistivity. (c) $T$-dependence of the superconducting transition width, $\Delta B_{c2} (= B_{c2}^\| - B_{c2}^\perp)$, for $B \parallel ab$ and $B \parallel c$. The dashed curves are fits for $T = 0$ extrapolation.

FIG. 3: (Color online) $B_{c2}$--$T_c$ phase diagram of EuFe$_2$As$_2$ for $B \parallel ab$ at 2.5 GPa. The values of $B_{c2}$ are determined under three different criteria, as illustrated for $\rho(B)$ at 4.2 K (inset). The solid or open symbols denote $B_{c2}$ determined from $\rho(B)$ and $\rho(T)$ measurements, respectively. The solid and dashed curves are fits to Eq. (1). $\times$ denotes the previous $B_{c2}^\|_0$ result deduced from an ac-$\chi$ measurement for $B \parallel ab$. The arrow indicates $T_N$ of Eu$^{2+}$ moments in the superconducting state in the absence of an applied field at 2.6 GPa

FIG. 4: (Color online) (a) $B_{c2}$--$T_c$ phase diagram of EuFe$_2$As$_2$ for $B \parallel c$ at 2.5 GPa. The solid and open symbols denote $B_{c2}$ deduced from $\rho(H)$ and $\rho(T)$ data, respectively. The dashed curves are fits to Eq. (1). The inset shows $B_{c2}^\|_0$ vs $T_c$ for $B \parallel ab$ and $B \parallel c$. The dashed curves are fits for $T = 0$ extrapolation.
son revealed that the obtained $\lambda_{ab}$ is comparable to that found in the Chevrel-type Eu compounds$^{32,35}$ and $B_{c}^{7}$ in EuFe$_2$As$_2$ is a few times greater than that reported in the Chevrel-type Eu compounds$^{32,35}$ We note that the concave curvature of $B_{c}$ in EuFe$_2$As$_2$ essentially differs from the positive curvatures often observed in highly two-dimensional SCs such as high-$T_c$ cuprates. In the latter, the curvature is highly dependent on what criterion is chosen to define $B_{c}$, and it is most likely affected by the vortex lattice phase transitions, i.e. from a vortex-liquid state to a vortex-solid state.$^{34}$

Figure 2(a) shows the $B_{c2}$-$T_c$ phase diagram of EuFe$_2$As$_2$ for $B \parallel c$ at 2.5 GPa,$^{20}$ determined in the same manner as that used for $B_{c2}^{ab}$. A concave curvature around 20 K is also visible for the $B_{c2}'$ curves. The dashed curves are calculated using the parameters comparable to those used for $B_{c2}^{ab}$, i.e., for $B_{c2}^{0}$, the fit gives ($\alpha$, $\lambda_{c2}$) = (1.9, 2.6) when we assume ($\beta$, $T_c$) = (-83, 24 K), identical to the values used for $B_{c2}^{ab}$. The calculated curves tend to saturate below 3 K, whereas the experimental curves appear to increase linearly as $T$ decreases to zero. The unsaturation of $B_{c2}'$ has been observed in other Fe-based SCs,$^{14,40–42}$ and it has been explained using a two-band model. Figure 2(b) shows the anisotropy ratio, $\Gamma = B_{c2}' / B_{c2}$, calculated from $B_{c2}^{0}(T)$ and $B_{c2}'(T)$. In spite of the quasi-two-dimensional layered structure in EuFe$_2$As$_2$, we obtain a small value of $\Gamma$, ranging between 0.9 and 1.4, which is comparable to that obtained for other “122” compounds.$^{20}$ In contrast to the monotonic decrease in $\Gamma$ with decreasing $T$ in other “122” compounds, $\Gamma$ in EuFe$_2$As$_2$ exhibits a broad maximum at around 8 K, which is likely ascribed to the presence of the $B_f$.

In order to compare the magnitude of $B_{c2}(0)$ with that of other Fe-based SCs, we estimate it by extrapolating the low-$T$ data to $T = 0$, as shown by the dashed curves in the inset of Fig. 2(a). For the extrapolations, an empirical expression, $B_{c2}(t) = B_{c2}(0)(1 - t^2)/(1 + t^2)$ and a linear fit are used for $B_{c2}^{ab}$ and $B_{c2}'$, respectively. We obtain $B_{c2}^{ab}(0) = 24.7$ T and $19.7$ T and $B_{c2}'(0) = 21.5$ T and 17.2 T for $B_{c2}^{ab}$ and $B_{c2}'$, respectively. $B_{c2}(0)$ in EuFe$_2$As$_2$ is significantly lower than $B_{c2}(0) > 50$ T in other Fe-based SCs at $T_c = 20 - 30$ K.$^{32,35}$ The width of the superconducting transition, $\Delta B (= B_{c2} - B_{c2}^{0})$, increases as $T$ decreases to 15 K for both $B \parallel ab$ and $B \parallel c$ [Fig. 2(c)]. Below 15 K, $\Delta B$ is virtually $T$-independent, as reflected by the parallel-shifts of the $\rho(B)$ curves in Fig. 2(a) and (c). The $T$-dependence may correlate with the development of $M$; $M$ at $B = B_{c2}(T)$ increases rapidly as $T$ decreases from $T_c$, but it is virtually saturated below $\sim 15$ K. At 1.6 K, $\Delta B$ is estimated as 5.1 T and 4.4 T for $B \parallel ab$ and $B \parallel c$, respectively. The relatively narrow transition width at low $T$, which is also observed in Ba(Fe,Co)$_2$As$_2$,$^{42,43}$ signifies a strong vortex pinning force in EuFe$_2$As$_2$.

In conclusion, we carried out high-field resistivity measurements up to 27 T for EuFe$_2$As$_2$ at 2.5 GPa, and we constructed the $B_{c2}$-$T_c$ phase diagram down to a minimum temperature of 1.6 K. Our analysis was based on a multiple pair-breaking model, and it revealed that the distinctive $B_{c2}$ curves with positive curvature and the reduced $B_{c2}$ values can be attributed to the substantial negative exchange field from the Eu$^{2+}$ moments. The low temperature anisotropy at 1.6 K, $\Gamma = 1.2$, is comparable to the results obtained for other “122” systems.

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1. Y. Kamihara et al., J. Am. Chem. Soc. 130, 3296 (2008).
2. For recent reviews, see K. Ishida et al., J. Phys. Soc. Jpn. 78, 062001 (2009) and D. C. Johnston, Advances in Physics 59, 803 (2010).
3. H. Kito et al., J. Phys. Soc. Jpn. 77, 063707 (2008).
4. Z.-A. Ren et al., Chin. Phys. Lett. 25, 2215 (2008).
5. C. Wang et al., Europhys. Lett. 83, 67006 (2008).
6. P. L. Alireza et al., J. Phys.: Condens. Matter 21, 012208 (2008).
7. Z. Ren et al., Phys. Rev. Lett. 102, 137002 (2009).
8. K. Matsubayashi et al., J. Phys. Soc. Jpn. 78, 073706 (2009).
9. T. Terashima et al., J. Phys. Soc. Jpn. 78, 083701 (2009); 78, 118001 (2009); Physica C 470, S443 (2010).
10. W. Yu et al., Phys. Rev. B 79, 020511(R) (2009).
11. T. Yamazaki et al., Phys. Rev. B 81, 224511 (2010).
12. N. R. Werthamer et al., Phys. Rev. 147, 295 (1966).
13. H. Kogawa et al., J. Phys. Soc. Jpn. 78, 013709 (2009).
14. F. Hunte et al., Nature 453, 903 (2008).
15. H. Raffius et al., J. Phys. Chem. Solids 54, 135 (1993)
16. H. S. Jevean et al., Phys. Rev. B 78, 052502 (2008).
17. Z. Ren et al., Phys. Rev. B 78, 052501 (2008).
18. Y. Xiao et al., Phys. Rev. B 80, 174424 (2009)
19. S. Jiang et al., N. J. Phys. 11, 025007 (2009)

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20. C. F. Miclea et al., Phys. Rev. B 79, 212509 (2009).
21. N. Kurita et al., arXiv: 1008.0684 (2010), to appear in J. Phys.: Conf. Ser. (2011).
22. K. Matsubayashi et al., arXiv:1007.2889 (2010).
23. T. Terashima et al., J. Phys. Soc. Jpn. 79, 103706 (2010).
24. Y. Xiao et al., Phys. Rev. B 81, 292046 (R) (2010)
25. Y. Uwatoko et al., Physica C 329-333, 1658 (2003)
26. K. Murata et al., Rev. Sci. Instrum. 79, 085101(2008)
27. R. H. Liu et al., Phys. Rev. Lett. 101, 087001 (2008).
28. M. Gooch et al., Phys. Rev. B 79, 104504 (2009).
29. N. Kurita et al., unpublished.
30. Ø. Fischer et al., J. Phys. C 8, L474 (1975).
31. M. S. Torikachvili and M. B. Maple, Solid State Commun. 40, 1 (1981).
32. M. Decroux et al., Phys. Rev. Lett. 52, 1563 (1984)
33. V. Jaccarino and M. Peter, Phys. Rev. Lett. 9, 290 (1962).
34. O. Fisher, Helv. Phys. Acta 45, 331 (1972).
35. C. Rossel et al., J. Appl. Phys. 57, 3099 (1985)
36. B. S. Chandrasekhar, Appl. Phys. Lett. 1, 7 (1962); A. M. Clogston, Phys. Rev. Lett. 9, 266 (1962)
37. The model reproduces experimental c-axis magnetization satisfactorily. The anisotropy of magnetization is small, except for a low-$T$ and low-$B$ region hence, we use the
same model for both $B \parallel ab$ and $B \parallel c$.

38 Y. Ando et al., Phys. Rev. B 60, 12475 (1999).

39 There is discrepancy between the present and previous $B_c^2$ data. The latter was determined from $\chi_9$. This could be due to the difference in the applied pressure and/or sample variations. In this study, $\rho$ was simultaneously measured using the two samples from the same piece.

40 H. Q. Yuan et al., Nature 457, 565 (2009).
41 S. A. Baily et al., Phys. Rev. Lett. 102, 117004 (2009).
42 M. Kano et al., J. Phys. Soc. Jpn. 78, 084719 (2009).
43 A. Yamamoto et al., Appl. Phys. Lett. 94, 062511 (2009).
44 A. Leitner et al., Phys. Rev. B 62, 1408 (2000).