PHYSICAL REVIEW B **94**, 020502(R) (2016)

Absence of superconductivity in the collapsed tetragonal phase of KFe_2As_2 under hydrostatic pressure

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(Received 7 May 2016; revised manuscript received 20 June 2016; published 15 July 2016)

Temperature dependence of resistivity on KFe_2As_2 single crystals down to 20 mK was measured under various hydrostatic pressures up to 17.5 GPa generated in a cubic-anvil cell. With increasing the pressure, the superconducting transition of tetragonal KFe_2As_2 was suppressed gradually and disappears completely at ∼11 GPa, which was related to the weakening of electronic correlations and/or critical fluctuations under pressure. In sharp contrast to previous reports, no superconducting phase emerges upon further increasing pressures until the collapsed tetragonal KFe_2As_2 forms. We argue that such a discrepancy can be attributed to the different pressure apparatus or homogeneity.

DOI: 10.1103/PhysRevB.94.020502

Superconductivity (SC) of $ThCr_2Si_2$-type tetragonal KFe_2As_2 has been studied intensively due to the controversy over the superconducting gap structure at ambient pressure [1–5]. More recently, a high-pressure (P) effect on the tetragonal KFe_2As_2 has attracted much attention [6–12] because of the observed unusual pressure dependence of superconducting transition temperature (T_c) from negative to positive around a critical pressure of $P_{c1} \sim 2$ GPa [6]. To interpret the "V"-shaped T_c under pressure, Tafti *et al.* proposed a change in the superconducting pairing symmetry from d to s waves at P_{c1}, whereas Taufour *et al.* argued a different mechanism as the development of a k_z modulation of the superconducting gap structures above P_{c1} [6,7]. The de Haas–van Alphen oscillations are in favor of a tiny modification of the Fermi surface and suggested a crossover from a nodal to a full-gap s wave at P_{c1} [8]. In addition, the high-pressure NMR study has convinced us that KFe_2As_2 is adjacent to an antiferromagnetic quantum critical point (QCP) at a negative pressure of −0.6 GPa and the decrease in T_c below 2 GPa was attributed to the suppression of spin fluctuations by applying pressure [12]. A magnetostriction and thermal expansion study also suggests a QCP at a negative pressure [13]. To date, the underlying mechanism of this reversal remains under hot debate.

With further increasing pressure, the tetragonal KFe_2As_2 at ambient pressure was found to transform into the collapsed tetragonal (cT) structure at a critical pressure of $P_{c2} \sim 16$ GPa at room temperature [9,10]. Interestingly, Nakajima *et al.* observed an abnormal drop of resistivity in the collapsed tetragonal KFe_2As_2 with a maximal characteristic temperature of ∼11 K, attributed to a superconducting transition from its field dependence [9]. In striking contrast, Ying *et al.* reported that the superconducting regions cover not only the collapsed tetragonal phase above 16 GPa, but also the high-pressure tetragonal phase over 10 GPa [10]. Based on these experiments, theoretical calculations proposed a pressure-induced Lifshitz transition associated with the collapsed tetragonal phase and the superconducting state [11]. However, the high-pressure SC phase diagram remains controversial because of the strong sensitivity of T_c depending on the type of the pressure cells and the pressure transmitting medium used as illustrated in Fig. 1. In addition, zero resistivity states cannot be observed under pressures above 7 GPa [9,10]. Especially, the superconductivity of the collapsed tetragonal KFe_2As_2 is an open question since there are no zero resistivity states in the previous studies [9,10] presumably due to the pressure inhomogeneity or the nonhydrostaticity. To resolve these issues pertaining to this intriguing compound, a high-pressure study on KFe_2As_2 under much improved hydrostatic pressure conditions is highly desirable. In this regard, the cubic-anvil cell (CAC) is a suitable high-pressure apparatus since it can generate nearly hydrostatic pressures even beyond 10 GPa [14,15]. In this Rapid Communication, we have performed resistivity measurements on KFe_2As_2 single crystals by using cubic-anvil cells up to 17.5 GPa. We found that the superconducting transition of tetragonal KFe_2As_2 was suppressed gradually and disappears completely around 11 GPa. In striking contrast with the previous studies [9,10], the collapsed tetragonal KFe_2As_2 does not show superconducting behavior. Our results suggest that the observation of the superconducting phase in the collapsed tetragonal KFe_2As_2 in previous reports should originate from the pressure inhomogeneity.

High quality KFe_2As_2 single crystals were grown with a flux method as described elsewhere [16]. The experiments under pressure were preformed in a cubic-anvil apparatus, which generates much improved hydrostatic pressures owing to the multiple-anvil geometry [14,15]. A preheated MgO cube was used as a gasket, and Daphne oil 7373 was used as the pressure transmitting medium. Resistivity was collected by a dc four-probe method with the current applied within the ab plane. Two samples No. 1 and No. 2 were studied: No. 1 was measured in a cubic-anvil-cell apparatus with a ^{4}He refrigerated chamber (2 K $\leqslant T \leqslant$ 300 K); No. 2 was measured in a clamp-type cubic-anvil cell with a ^{3}He /^{4}He dilution refrigerator (BF-LD400, 20 mK $\leqslant T \leqslant$ 300 K).

Figure 1 shows the superconducting phase diagram and the superconducting transition widths of KFe_2As_2 under pressure

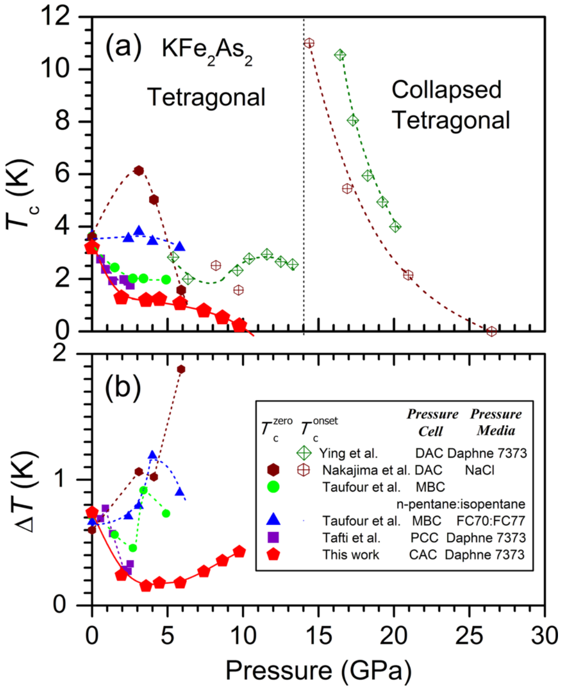

FIG. 1. (a) Temperature-pressure phase diagram of KFe_2As_2 and the comparisons with the previous data. The solid and open symbols represent the zero resistivity state temperature T_c^{zero} and the onset temperature T_c^{onset} of the superconducting transition, respectively. (b) $\Delta T_c (= T_c^{onset} - T_c^{zero})$ as a function of pressure. (DAC), (MBC), (PCC), (CAC) represent diamond-anvil cell, modified Bridgeman cell, piston cylinder cell, and cubic-anvil cell, respectively.

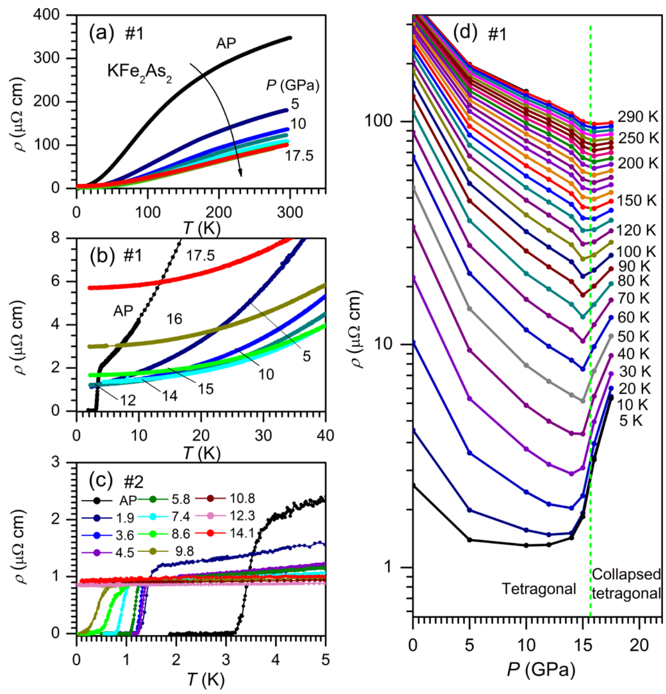

FIG. 2. (a) Temperature dependence of resistivity ρ for No. 1 up to 17.5 GPa; (b) the enlarged low-T data. (c) ρ for No. 2. (d) ρ under pressure at fixed temperatures for No. 1.

together with the previous reported data for comparison. At ambient pressure, T_c^{zero} is $\sim$3.2 K with the superconducting transition widths $\Delta T_c \sim 0.7$ K (ΔT_c defined as the differences of zero resistivity temperature T_c^{zero} and the onset temperature T_c^{onset}), consistent with the reported values [6,7,17]. As the pressure increases, T_c shifts quickly down to 1.3 K at 1.9 GPa and then increases slightly for $3.6 < P < 4.5$ GPa before decreasing again for $P > 4.5$ GPa. At $\sim$11 GPa, the superconducting state of tetragonal KFe_2As_2 disappears completely. The pressure dependence of T_c obtained in this Rapid Communication is generally in line with the previous data under hydrostatic pressure, but several distinct features are evidenced: first, ΔT_c under pressure becomes much narrower than the reported values in the same pressure region. As the pressure increases, in Fig. 1(b), ΔT_c decreases quickly and is only $\sim$0.2 K for $1.9 < P < 5.8$ GPa, comparable with the data under better hydrostatic conditions below 2.5 GPa [6,7]. In this Rapid Communication, the zero resistivity state retains up to 11 GPa before the superconducting state disappears completely. Differently, in other reports, the superconducting transition becomes broad evidently as the pressure increases and ΔT increases to $\sim$0.5–2 K depending on the pressure apparatus and/or pressure media used; above 7 GPa, the zero resistivity state cannot be achieved, and the superconducting transition always extends over a large temperature range [9,10]. These differences are attributed to different pressure conditions. Second, T_c obtained in this Rapid Communication is lower than others in the same pressure regions in Fig. 1(a). As proposed in the previous studies, T_c of tetragonal KFe_2As_2 depends sensitively on pressure conditions and was enhanced if under a less hydrostatic condition [7]. In this sense, the lower T_c observed in this Rapid Communication suggests a much improved hydrostatic pressure condition in a cubic-anvil cell in comparison with the PCC or DAC under higher pressure. At last, we examined the resistivity of KFe_2As_2 in the pressure region of $14 < P < 17.5$ GPa, however, superconductivity does not appear down to 2 K, which is contrary to the previous results using DAC [9,10]. The origin will be discussed later.

Figure 2 shows the temperature dependence of resistivity $\rho(T)$ under various pressures. At ambient pressure, KFe_2As_2 displays metallic behaviors and enters a superconducting state below 3.2 K. With increasing the pressure, resistivity at room temperature monotonously decreases up to 16 GPa and then shows a slight increase at high pressure. The critical pressure almost coincides with the reported value for the tetragonal to collapsed tetragonal structural transition determined by x-ray diffraction measurement under pressure using the DAC with helium or argon as the pressure media [9,10]. To see this point more clearly, we replotted the resistivity vs pressure at fixed temperatures in Fig. 2(d). As seen, resistivity vs pressure displays a V-type shape with a minimum at a critical pressure. Next, we examined the resistivity above 11 GPa in Fig. 2(b). Low-temperature ρ increases continuously on the warming process, and no drop emerges. As shown in Fig. 2(c), ρ of No. 2 was measured down to 20 mK. With increasing the pressure, the superconducting state of tetragonal KFe_2As_2 was suppressed and completely disappeared at 11 GPa. In Fig. 4(a), both T_c^{onset} and T_c^{zero} decrease as the pressure increases, and ΔT_c shows a "trough"-type behavior with a minimum of 0.16 K at 3.6 GPa and then starts to increase

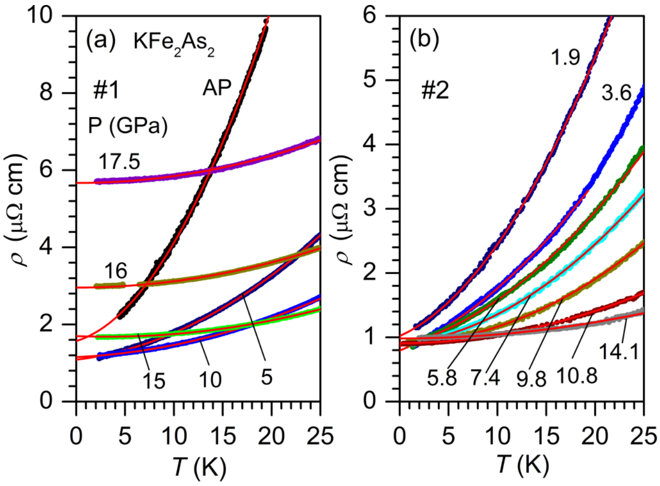

FIG. 3. The ρ data were fitted by using the formula $\rho = \rho_0 + A_1T + A_2T^2$, the residual resistivity ρ_0, and the coefficients A_1, and A_2.

above 7.4 GPa. As the pressure increases up to the region of $11 \leqslant P \leqslant 14.1$ GPa, no superconducting transition appear down to 20 mK. Combined with the above results, we reach the conclusion that the collapsed tetragonal KFe_2As_2 is not superconducting.

Normal-state resistivity close to T_c provides useful information about superconductivity. At first, we fitted the ρ data by using $\rho = \rho_0 + AT^\alpha$ up to 14 K where ρ_0 represents the residual resistivity, the temperature coefficient A, and the exponent α. The exponent α was found to be 1.7 ± 0.2 at ambient pressure and shows weak pressure dependence. On the other hand, as the pressure increases up to higher than 11 GPa, α reaches up to ∼2, indicating the Fermi-liquid state. Therefore, a gradual crossover from non-Fermi-liquid to Fermi-liquid behavior appears under pressure. There is a suggestion that the non-Fermi-liquid behavior at ambient pressure could be explained by multiband effects [17]. However, the multiband analysis is very complicated, and the simple fitting of resistivity may not be enough because one should examine whether the carrier scattering times for different carriers obey Fermi-liquid T^2 behavior. In this Rapid Communication, we adopt instead an empirical formula $\rho = \rho_0 + A_1T + A_2T^2$ [18] to construct a qualitative relation between the evolution of temperature coefficient and the T_c. Here, the T^2 term is to describe the Fermi-liquid state, and the T-linear term is associated with the electronic correlations and scattering process, such as the electron-boson interaction and/or critical fluctuations near QCP [18,19]. The results of fitting and the obtained parameters were presented in Figs. 3 and 4, respectively. The residual resistivity ρ_0 decreases monotonically, starts to increase at 14 GPa, then jumps to nearly three times with the pressure increasing up to 17.5 GPa, which is sharply different from the reports by using DAC [9]. In Ref. [9], ρ_0 keeps increasing monotonously and then reaches a nearly constant in the collapsed tetragonal phase, which indicates the tetragonal to collapsed tetragonal transition covers a wide pressure region. This character is clear evidence of the nonhydrostatic pressures or the wide pressure distributions in DAC using NaCl as a pressure medium. As the pressure increases, A_1 decreases linearly and becomes

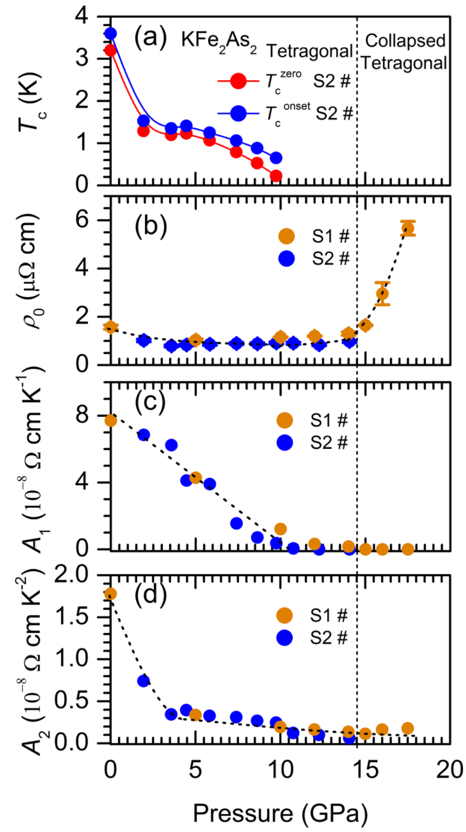

FIG. 4. The evolutions of parameters under pressure: (a) T_c^{onset} and T_c^{zero}, (b) ρ_0, (c) A_1, and (d) A_2. The dashed lines indicate phase transformation at lower temperatures.

almost zero around 11 GPa, coinciding with the suppression of T_c. This close connection between T_c and A_1 suggests that the scattering mechanism leading to the T-linear term plays an important role for the appearance of SC in KFe_2As_2. On the other hand, A_2 decreases rapidly with approaching P_{c1} and then gradually decreases with further increasing pressure, which is consistent with the previous report [7]. Furthermore, this observation is in agreement with a magnetic quantum critical point located at a negative pressure [12,13]. Incidentally, the shallow minimum of T_c near ∼2 GPa is not explicable in terms of the pressure dependence of A_1 and A_2. Other factors, such as pressure variation of the density of state at the Fermi level, balance between intra- and inter-Fermi-surface-pocket scatterings, would also affect T_c and its pressure dependence. The combined interplays of those factors would be important to fully understand the evolution of T_c under pressure.

Finally, we discuss the absence of superconductivity in collapsed tetragonal KFe_2As_2. It is generally accepted that pressure is a clean method to fine-tune the electronic structure. However, it is complicated if there exists an inhomogeneous pressure distribution. This effect becomes more serious in the pressure cells with a uniaxial geometry (e.g., PCC, DAC, or the modified Bridgeman cell) due to the solidification of the liquid pressure medium or the direct use of a solid pressure-transmitting medium, such as NaCl. The situation becomes important for the layered structures. For example, the superconducting state in $BaFe_2As_2$ and $SrFe_2As_2$ emerges at a critical pressure of ∼3 GPa under an uniaxial pressure in DAC [20], much lower than those in cubic-anvil cells with better hydrostatic conditions (~10 GPa in $BaFe_2As_2$ and ~5 GPa in $SrFe_2As_2$) [21,22]. In less hydrostatic pressure conditions, the actual pressures are changing along different crystal axes, which cause the discrepant pressure dependence of T_c in anisotropic $BaFe_2As_2$ or $SrFe_2As_2$. These examples illustrate severe influence of the pressure distribution or nonhydrostaticity especially near the phase boundary for a structural transition [23,24]. The strong sensitivity to nonhydrostatic pressure in 122-type iron-based superconductors suggests that a high-pressure experiment under hydrostatic conditions is required to obtain the intrinsic properties. In previous reports, the collapsed tetragonal KFe_2As_2 was reported to exhibit superconductivity by using a DAC with a pressure medium of NaCl and liquid Daphne oil 7373 [9,10]. In this Rapid Communication, the resistivity was collected by using the cubic-anvil cell. As above, the lower T_c and smaller width ΔT_c were achieved, and the zero resistivity state retains up to 11 GPa, which can be seen as evidence of good hydrostatic pressures in comparison to the previous studies [9,10]. Moreover, to eliminate the effect of a pressure medium, liquid Daphne oil 7373 was chosen, which is the same as used in the reports using DAC [10]. However, no SC was found in the collapsed tetragonal KFe_2As_2 and the tetragonal KFe_2As_2 above 11 GPa. As we know, the solidification pressure of Daphne oil 7373 at room temperature is ~2.2 GPa, above which the uniaxial pressure is inevitable especially in DAC, whereas the quasihydrostatic condition is maintained owing to the multianvil geometry in the CAC. We also note the fact that the reported superconductive regions in the collapsed tetragonal phase of KFe_2As_2 are proximate to the first-order structural transition and there is no zero resistivity state nor diamagnetic susceptibility. Accordingly, we suggest that the reported drop in resistivity of the collapsed tetragonal KFe_2As_2 [9,10] is not a superconducting transition but possibly related to nonhydrostatic pressure effects, such as the uniaxial-strain-induced phase separations by the solidifications of pressure media. The previously reported superconductivity of the collapsed tetragonal KFe_2As_2 by using DAC, however, is absent in the cubic-anvil pressure cell, and future theoretical and experimental studies are required to clarify.

To summarize, the superconducting transition of the tetragonal KFe_2As_2 was suppressed gradually and disappears completely at ~11 GPa. No superconductivity appears in the collapsed tetragonal phase of KFe_2As_2. The discrepancy between the present and the previous reports probably originates from the difference in pressure homogeneity.

We thank S. Nagasaki for technical assistance. B.W. and Y.U. acknowledge support of JSPS fellowships (Grants No.15F15023 and No.15H03681). J.C. is supported by the National Science Foundation of China (Grant No. 11574377) and the Strategic Priority Research Program of the Chinese Academy of Sciences (Grant No. XDB07020100). C.-H.L was supported by a Grant-in-Aid for Scientific Research B (Grant No. 24340090) from the Japan Society for the Promotion of Science.

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