Absence of superconductivity in the collapsed tetragonal phase of KFe$_2$As$_2$ under hydrostatic pressure

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Temperature dependence of resistivity on KFe$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ forms. We argue that such a discrepancy can be attributed to the different pressure apparatus or homogeneity.

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Superconductivity (SC) of ThCr$_2$Si$_2$-type tetragonal KFe$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ single crystals by using cubic-anvil cells up to 17.5 GPa. We found that the superconducting transition of tetragonal KFe$_2$As$_2$ was suppressed gradually and disappears completely around 11 GPa. In striking contrast with the previous studies [9,10], the collapsed tetragonal KFe$_2$As$_2$ does not show superconducting behavior. Our results suggest that the observation of the superconducting phase in the collapsed tetragonal KFe$_2$As$_2$ in previous reports should originate from the pressure inhomogeneity.

High quality KFe$_2$As$_2$ 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 $^3$He refrigerated chamber (2 K $\leq T \leq 300$ K); No. 2 was measured in a clamp-type cubic-anvil cell with a $^3$He/$^4$He dilution refrigerator (BF-LD400, 20 mK $\leq T \leq 300$ K).

Figure 1 shows the superconducting phase diagram and the superconducting transition widths of KFe$_2$As$_2$ under pressure.
FIG. 1. (a) Temperature-pressure phase diagram of KFe$_2$As$_2$ and the comparisons with the previous data. The solid and open symbols represent the zero resistivity state temperature $T_{c\text{zero}}$ and the onset temperature $T_{c\text{onset}}$ of the superconducting transition, respectively. (b) $\Delta T_c(=T_{c\text{onset}}-T_{c\text{zero}})$ as a function of pressure. (DAC), (MBC), (PCC), (CAC) represent diamond-anvil cell, modified Bridgeman cell, piston cylinder cell, and cubic-anvil cell, respectively.

As proposed in the previous studies, $T_c$ of tetragonal KFe$_2$As$_2$ 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$_2$As$_2$ 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.

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above 7.4 GPa. As the pressure increases up to the region of \( 11 \leq P \leq 14.1 \) GPa, no superconducting transition appears down to 20 mK. Combined with the above results, we reach the conclusion that the collapsed tetragonal KFe\(_2\)As\(_2\) is not superconducting.

Normal-state resistivity close to \( T_c \) provides useful information about superconductivity. At first, we fitted the \( \rho \) data by using \( \rho = \rho_0 + A_1T + A_2T^2 \), the residual resistivity \( \rho_0 \), and the coefficients \( A_1 \), and \( A_2 \).

The exponent \( \alpha \) 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, \( \alpha \) 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 \( \rho_0 \) 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. \([9]\), \( \rho_0 \) 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 almost zero around 11 GPa, coinciding with the suppression of \( T_c \).

Finally, we discuss the absence of superconductivity in collapsed tetragonal KFe\(_2\)As\(_2\). 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\(_2\)As\(_2\) and SrFe\(_2\)As\(_2\) 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$_2$As$_2$ and ∼5 GPa in SrFe$_2$As$_2$) [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$_2$As$_2$ or SrFe$_2$As$_2$. 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$_2$As$_2$ 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 $\Delta T_c$ 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$_2$As$_2$ and the tetragonal KFe$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ [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$_2$As$_2$ 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$_2$As$_2$ was suppressed gradually and disappears completely at ∼11 GPa. No superconductivity appears in the collapsed tetragonal phase of KFe$_2$As$_2$. The discrepancy between the present and the previous reports probably originates from the difference in pressure homogeneity.

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