Superconductivity in pressurized Rb$_{0.8}$Fe$_{2-y}$Se$_{2-x}$Te$_x$

Dachun Gu$^1$, Qi Wu$^1$, Yazhou Zhou$^1$, Peiwen Gao$^1$, Jing Guo$^1$, Chao Zhang$^1$, Shan Zhang$^1$, Sheng Jiang$^3$, Ke Yang$^3$, Aiguo Li$^3$, Liling Sun$^{1,2}$ and Zhongxian Zhao$^{1,2}$

1 Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100190, People’s Republic of China
2 Collaborative Innovation Center of Quantum Matter, Beijing, 100190, People’s Republic of China
3 Shanghai Synchrotron Radiation Facilities, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201204, People’s Republic of China

E-mail: llsun@iphy.ac.cn and zhxzhao@iphy.ac.cn

Keywords: alkaline iron selenide superconductor, superconducting phase diagram, pressure effect

Abstract
We report the finding of pressure-induced elimination and reemergence of superconductivity in Rb$_{0.8}$Fe$_{2-y}$Se$_{2-x}$Te$_x$ ($x = 0, 0.19$ and $0.28$) superconductors that belong to the family of A-245 superconductors ($A = K, Rb, TlRb$ and Cs), characterized by the presence of an antiferromagnetic (AFM) long-ranged order phase with the superlattice structure of Fe-cavities. In this study, we investigate the connections between superlattice, AFM phase and superconductivity via the combined approaches of Te doping and application of external pressure. Our data reveal that the superconductivity of the ambient-pressure superconducting phase (SC-I) and the AFM long-ranged order as well as the superconductivity of the pressure-induced phase (SC-II) in the host samples can be synchronously tuned by Te doping. At $x = 0.4$, the SC-I and AFM long-range ordered phases as well as the SC-II phase disappear together, indicating that the two superconducting phases have intrinsic connections with the AFM phase. Furthermore, in-situ synchrotron x-ray diffraction measurements indicate that the superlattice structure in the $x = 0.4$ sample still exists at ambient pressure, but collapses at the same pressure where the superlattice of the superconducting samples is destroyed. These results provide new insight into understanding the physics of this type of superconductors.

1. Introduction

The discovery of superconductivity in A$_{0.8}$Fe$_{2-x}$Se$_2$ compounds ($A = K, TlRb$) [1, 2] with ordered Fe vacancies, featured by $\sqrt{5} \times \sqrt{5}$ arrangement (defined as A-245 superconductors thereafter), provides a new platform to investigate connections among superconductivity, antiferromagnetic (AFM) ordered state and lattice structure in Fe-based superconductors [3–9]. Soon after, the superconductivity was also found in Rb-245 and Cs-245 compounds [10–15]. Then, the characteristics of lattice, electronic and magnetic structures have been widely reported for these superconductors, such as the existence of phase separation [16–19], the superlattice of ordered Fe vacancies and its connection to the AFM ordered phase [20–23], absence of hole pockets at the Fermi surface [24–26], temperature-induced orbital selection [27–29]. All of these features are shown neither in the iron arsenide nor in copper oxide superconductors, therefore the complexity of understanding its superconducting mechanism is raised to a new level.

Previous studies found that applying external pressure on the A$_{0.8}$Fe$_{1.5}$Se$_2$ ($A = K, Rb$ and Cs) superconductors can fully suppress the superconductivity of the ambient-pressure superconducting phase (SC-I) [30–33] and induce a new superconducting phase (SC-II) at higher pressure [33]. Experimental evidences have exhibited that the pressure-induced SC-II phase is probably driven by a quantum critical phase transition in which the AFM phase in the pressurized sample undergoes a transition from an AFM state to a paramagnetic (PM) state [30, 34]. These results suggest that the superconductivity of the SC-II phase may be closely related to the AFM fluctuations [35, 36]. On the other hand, it is known that the iso-valence substitution of Te or S (with larger or smaller ionic radius) for Se in the alkaline iron selenide superconductor can distort its
lattice, which can result in a suppression of the long-ranged AFM order and the superconductivity in the SC-I phase [37–40], but cannot induce a SC-II phase. The pressure-induced reemergence of superconductivity in A0.8Fe1.5Se2 (A = K and TlRb) superconductors attracts considerable attention [41–44], meanwhile, puzzles such as whether the SC-I and SC-II phases are intrinsically connected to each other and what the connections are among the superconductivity, AFM phase and superlattice are raised. The answers for these questions may be helpful to shed light on understanding the superconducting mechanism for this family of Fe-based superconductors. In this study, we combine the two tuning ways, doping Te on Se sites and the application of external pressure, to conduct comprehensive investigations on Rb-245 superconductors.

2. Experimental details

Single crystals of Rb-245 superconductors were grown out by the self-flux method as reported in [37]. The actual chemical compositions of all samples investigated were Rb0.8Fe1.63Se1.72Te0.28 and Rb0.8Fe1.66Se1.6Te0.4, respectively, which are identified by the inductive coupled plasma-atomic emission spectrometer.

In-situ high-pressure electrical resistance and ac susceptibility measurements were carried out in a non-magnetic diamond anvil cell which is integrated into a home-built refrigerator. Diamond anvils of 500 and 300 μm flats were used for this study. High-pressure resistance measurements were performed by a standard four-probe method in a diamond-anvil cell. Platinum electrodes with dimensions of 20 μm in width and 2.5 μm in thickness were used. The crystal was placed into the hole of the isolating gasket assembled with four electrodes, all of which were put on an anvil. And then the other anvil was pressed. To achieve a quasi-hydrostatic pressure condition, the NaCl powder was employed as the pressure medium for the resistance measurements. High-pressure ac susceptibility measurements were conducted by using home-made coils which were set up around the diamond anvils [33, 45]. Before sample loading, we performed susceptibility measurements for the high pressure cell with the coils and gasket only, and took the result as a background signal. The high-pressure magnetic susceptibility data were extracted through the background subtraction [45, 46]. Temperature was measured with a calibrated Si-diode attached to the diamond anvil cell with an accuracy less than 0.1 K. High-pressure x-ray diffraction (XRD) experiments were performed at beam line 15U at the Shanghai Synchrotron Radiation Facility (SSRF). Diamonds with low birefringence were selected for the XRD experiments. A monochromatic x-ray beam with a wavelength of 0.6199 Å was adopted for all measurements. Pressure was determined by the ruby fluorescence method [47].

3. Results and discussions

Figure 1(a) shows the resistance (R) as a function of temperature (T) for the undoped Rb-245 superconductor measured at different pressures. It can be seen that the R–T curve demonstrates a remarkable hump around 200 K. The resistance hump is suggested to have originated from the competition between the insulating AFM phase and the superconducting phase [27, 28, 36]. Upon increasing pressure, the hump is suppressed significantly, same as that seen in the pressurized K-245 and Tl(Rb)-245 superconductors [30, 32]. At pressure ~8.4 GPa, we found that the resistance hump becomes almost featureless, which is signed the destruction of the long-ranged AFM order [34]. Zooming in the plot of R–T curve in the low temperature range, the pressure-induced decrease in Tc is shown more clearly (figure 1(b)). At 7.2 GPa, the superconductivity is fully suppressed, and then the pressure-induced resistance drop is visible again in the pressure range from 8.4 to 11.8 GPa (figure 1(c)). On further increasing of pressure to 14.1 GPa, this resistance drop vanishes, similar to that seen in other A-245 superconductors [33]. To fully characterize the superconducting state in the pressurized Rb-245 superconductor, we performed ac susceptibility measurements at pressures of 1.1, 3.5 and 11 GPa, respectively. The results show that the host sample at these three pressure points is diamagnetic, indicating that the sample is superconducting (figures 1(d) and (e)). We repeated the measurements three times and found that the data are reproducible. Assuming the superconducting volume fraction of the SC-I phase in the Rb0.8Fe1.5Se2 superconductor at 1.1 GPa is 100% (magnitude of its superconducting transition measured from the real part of the susceptibility measurements is about 42 nV), our high-pressure ac susceptibility result (11.3 nV) indicates that the superconducting volume fraction of the SC-II phase is about 26.9% at 11 GPa. Further resistance measurements under magnetic field or dc current for the sample subjected to 8.4 GPa find that the R–T curves shift to lower temperature when the magnetic field or dc current is increased (figures 1(f) and (g)), providing further evidence for the existence of a pressure-induced SC-II phase.

Next we performed high-pressure studies on the Te-doped Rb-245 superconductors. We find that the resistance hump also exists in the pressure-free samples Rb0.8Fe1-xSe2-xTe_x (x = 0.19 and 0.28) (figures 2(a) and (f)). Applying external pressure yields a dramatic suppression on the resistance hump in these two samples. After
Careful inspection of their $R$-$T$ plots in the lower temperature range, we find that the $T_c$ of the SC-I phase declines with increasing pressure (figures 2(b) and (g)). Upon further increasing pressure, the resistance drops featuring the SC-II phase show up at 11.5 GPa for the $x=0.19$ sample and at 12.4 GPa for the $x=0.28$ sample, respectively (figures 2(c) and (h)). The superconducting transition of the SC-II phase in these two samples is confirmed by the shift of the $R$-$T$ curve to a lower temperature when the magnetic field or current is increased (figures 2(d), (e), (i) and (j)).

Figure 3 illustrates the temperature dependence of resistance for the $x=0.4$ sample at different pressures, and notably the sample at ambient pressure is in a semiconducting state. With increasing pressure, the semiconducting behavior is suppressed dramatically (figures 3(b) and (c)). At 13 GPa and above, its resistance decreases remarkably with decreasing temperature (figure 3(d)), indicating that the sample transforms into a metallic state. No SC-II phase is detected in the pressurized sample up to 15.5 GPa (figure 3(e)).

The overall behavior of Rb-245 superconductors is summarized in the electronic phase diagram of pressure-composition-temperature, as shown in figure 4. Adopting pressure as a control parameter, the $T_c$ of the SC-I phase in the $x=0$ sample decreases with increasing pressure, and a new superconducting phase (SC-II) emerges within 8.4 GPa–11.8 GPa, after the SC-I phase is fully suppressed. The maximum onset $T_c$ of the SC-II phase is $\sim 53$ K at 11.8 GPa. The diagram with double superconducting phases has been observed in K-245 and Tl(Rb)-245 superconductors [33], so the results reported in this study further indicate that the pressure-induced reemergence of superconductivity is a common phenomenon for the family of A-245 superconductors.

For $x=0.19$ and 0.28 samples, the ambient-pressure value of the $T_c$ in their SC-I phase is lower than that ($T_c=33$ K) of the undoped sample, $T_c=29.8$ K for the $x=0.19$ sample and 24.2 K for the $x=0.28$ sample (left panels of figure 4), implying that Te-doping is not in favor of superconductivity [37–39]. Remarkably, the reduced $T_c$ of the SC-I phase can be partially recovered by applying pressure, and the maximum recovered $T_c$ value for the $x=0.19$ sample at 1.2 GPa is 1.1 K and for the $x=0.28$ sample at 1.7 GPa is 2.2 K, respectively. Our results reveal that the $T_c$ of the SC-I phase in the A-245 superconductors is very sensitive to the local lattice distortion from Te doping.
It is noteworthy that neither the SC-I nor SC-II phases is observed in the sample of $x = 0.4$, at the doping level of which the long-ranged AFM order at ambient pressure is fully suppressed [30, 34, 36, 37] (upper right panel of figure 4). No observable sign exists of the SC-II phase, even when the paramagnetic semiconducting sample ($x = 0.4$) is pressurized into a metallic state (figures 3(d) and (e) and 4). Previous high-pressure studies on K-245 and TlR-245 superconductors revealed that the SC-II phase emerges from a metallic state, driven by a quantum critical transition [30]. While, because the $x = 0.4$ sample is in a paramagnetic semiconducting state, pressure is unable to turn on a SC-II phase from such a heavy-doped sample.

Neutron diffraction studies on A-245 superconductors have confirmed that the AFM long-ranged order is associated with the superlattice structure [34]. To clarify the role of the superlattice in stabilizing the superconductivity of the A-245 superconductors, we performed high-pressure synchrotron x-ray diffraction measurements at SSRF for Rb$_{0.8}$Fe$_{2-y}$Se$_{2-x}$Te$_x$ ($x = 0, 0.19, 0.28$ and $0.4$) samples. As shown in figure 5, we find that a superlattice peak (110) exists in all samples investigated below ~9 GPa. At pressure above ~10 GPa, the superlattice peak disappears from all these samples, consistent with the results observed in K$_{0.8}$Fe$_{1.78}$Se$_2$ and Tl$_{0.6}$Rb$_{0.4}$Fe$_{1.67}$Se$_2$ superconductors [30, 33, 34]. More significantly, we find that the superlattice peak of the $x = 0.4$ sample still exists at ambient pressure although its AFM long-ranged order state is fully suppressed [37]. This result demonstrates that Te doping can destroy the AFM order state by partially destructing the superlattice
Figure 3. Temperature dependence of resistance for Rb$_{0.8}$Fe$_{1.66}$Se$_{1.6}$Te$_{0.4}$ at different pressures. (a) R-T curves measured in the pressure range of 0.5–15.5 GPa, showing a pressure-induced remarkable suppression of the insulating behavior. (b)–(e) R-T curves measured at 2.2, 5.7, 14.2 and 15.5 GPa, respectively.

Figure 4. Phase diagram of temperature–doping–pressure of Rb–245 samples. Te doping suppresses the superconductivity of SC-I and SC-II phases. As the doping level reaches 0.4, SC-I and SC-II disappear together in the pressure range investigated, demonstrating an intimate connection between the two superconducting phases. The data of upper right panel is taken from [37]. The left panels display two-dimensional temperature–pressure phase diagrams for different Te dopings. The green and red dotted lines guide to eye.
structure, while applying pressure can result in the entire destruction of the superlattice structure [34], revealing important issues that the AFM long-range ordered state is sensitive to the local distortion of the superlattice structure induced by Te doping.

4. Conclusions

In this study, we find the pressure-induced reemergence of superconductivity in Rb$_{0.8}$Fe$_{2-y}$Se$_{2-x}$Te$_x$ ($x = 0, 0.19$ and $0.28$) superconductors and the connection between the SC-I and SC-II phases. Our results demonstrate that Te-doping can significantly suppress the superconductivity of the SC-I and SC-II phases, and eliminate these two superconducting phases and the AMF order at $x = 0.4$. We propose that the SC-I and SC-II phases are connected by the state of the AFM phase, i.e. the AFM long-range ordered state stabilizes the superconductivity of SC-I phase, while the pressure-induced AFM fluctuation state drives the reemergence of superconductivity of the SC-II phase. The superconducting phase diagram obtained in this study provides a panorama picture on the pressure and doping planes for the superconducting behaviors of the A-245 superconductors. Significantly, in-situ high-pressure x-ray diffraction measurements for all samples investigated reveal that the superlattice structure exists below $\sim$10 GPa for all samples.

Acknowledgments

We thank Prof. Jianqi Li for valuable discussions. This work has been supported by the NSF of China (Grant No. 91321207 and 11427805), 973 projects (Grant No. 2011CBA00100) and the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (Grant No. XDB07020300).

References

[1] Guo J G, Jin S F, Wang G, Wang S C, Zhu K X, Zhou T T, He M and Chen X L 2010 Phys. Rev. B 82 180520 (R)
[2] Fang M H, Wang H D, Dong C H, Li Z J, Feng C M, Chen J and Yuan H Q 2011 Europhys. Lett. 94 27009
[3] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[4] Ronning F, Klimczuk T, Bauer E D, Volz H and Thompson J D 2008 J. Phys.: Condens. Matter 20 322201
[5] Rotter M, Tegel M, Johrendt D, Schellenberg I, Hermes W and Pöttgen R 2008 Phys. Rev. B 78 020503 (R)
[6] Tegel M, Rotter M, Weiß V, Schappacher F, Pöttgen R and Johrendt D 2008 J. Phys.: Condens. Matter 20 452201
