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Coevolution of superconductivity and Hall coefficient with anisotropic lattice shrinkage in compressed $KCa_2Fe_4As_4F_2$

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The stability of superconductivity in superconductors is widely recognized to be determined by various factors, including charge, spin, orbit, lattice, and other related degrees of freedom. Here, we report our findings on the pressure-induced coevolution of superconductivity and Hall coefficient in $KCa_2Fe_4As_4F_2$, an iron-based superconductor possessing a hybrid crystal structure combining KFe_2As_2 and CaFeAsF. Our investigation, involving high-pressure resistance, Hall effect and x-ray diffraction (XRD) measurements, allows us to observe the connection of the superconductivity and Hall coefficient with the anisotropic lattice shrinkage. We find that its ambient-pressure tetragonal (T) phase presents a collapse starting at around 18 GPa, where the sign of the Hall coefficient (R_H) changes from positive to negative. Upon further compression, both superconducting transition temperature (T_c) and R_H exhibit a monotonous decrease. At around 41 GPa, the superconductivity is completely suppressed ($T_c = 0$), where the parameter a begins to decline again and the Hall coefficient remains nearly unchanged. Our experiment results clearly demonstrate that the pressure-induced anisotropic lattice collapse plays a crucial role in tuning the interplay among multiple degrees of freedom in the superconducting system and, correspondingly, the stability of the superconductivity.

Keywords: effects of pressure, pnictides and chalcogenides, transport properties

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The discovery of iron-based superconductors[1,2] provides a new opportunity for better understanding the mechanism of high-T_c superconductivity, a topic that has been the subject of debate for over three decades. Currently, various types of iron-pnictide superconductors with different structures have been identified. These types primarily include superconductors with stacking sequences of LaOFeAs (1111), Sr_2VO_3FeAs (21311), FeSe (11), $BaFe_2As_2$ (122), $CaKFe_4As_4$ (1144), and $KCa_2Fe_4As_4F_2$ (12442).[1,3–7] Among this kind of superconductors, there exists a distinct group that displays unique characteristics. Examples of such superconductors include $Ca_{10}(Pt_4As_8)(Fe_2As_2)_5$ (1048),[8] $Ba_2Ti_2Fe_2As_4O$ (22241),[9] 1144,[6] and 12442[7,10] superconductors, which have specific chemical compositions with stoichiometry. This allows them to exhibit intrinsic superconductivity without the requirement of chemical doping. The findings of these stoichiometric superconductors provide a remarkable platform for understanding the mechanism of the high-T_c superconductivity because they significantly reduce the complexity associated with the inhomogeneous local structure from chemical doping.

There is consensus that the stability of superconductivity of iron-based superconductors is highly related to the Fe–As coupling.[11,12] Notably, high pressure has proven to be an effective method for manipulating superconductivity and electronic structure.[13–24] In particular, the application of high pressure allows for the adjustment of the bond length within the Fe–As layer through lattice compression.[25] Recent high-pressure studies on the stoichiometric 1144 superconductors found that the transition from a half-collapse to a full-collapse of the tetragonal phase has a profound impact on the superconductivity[25–28] due to the reconstruction of Fermi surface.[25] In comparison to the 1144 superconductor, the 12442 superconductor possesses a more complex crystal structure.[7,29–31] It can be viewed as an intergrowth structure that combines the alternating layers of KFe_2As_2 and CaFeAsF.[7,10] This structure includes double Fe_2As_2 layers with K atoms sandwiched in between, and these layers are separated by a Ca_2F_2 layer. Consequently, the 12442 superconductor exhibits two different sites for As ions within the tetragonal unit cell, namely, As(1) and As(2), with a space group $I4/mmm$.[7,10] Upon cooling, $KCa_2Fe_4As_4F_2$ exhibits superconductivity with a critical temperature (T_c) around 33 K. Notably, angle-resolved photoemission spectroscopy and specific heat experiments have revealed that this material possesses a complex electronic

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structure and multigap properties,[29,32–34] consistent with the theoretical calculations.[35] However, the gap symmetry of $KCa_2Fe_4As_4F_2$ remains a subject of controversy.[32–34,36,37] Previous high-pressure studies conducted on the 12442 superconductor up to 15 GPa have demonstrated that the superconducting transition temperature (T_c) initially increases, reaches a maximum of $T_c = 36.5$ K at 2 GPa and then decreases upon further compression, and no iso-structural phase transition was observed within the investigated pressure range.[38] Given that the 12442 superconductor shares a similar structure to that of the 1144 superconductor, which demonstrates a close relationship between T_c, Hall coefficient, and lattice structure under pressure, it becomes highly intriguing to investigate whether the 12442 superconductor also exhibits the same correlation under higher pressure. This investigation aims to assess how the lattice shrinkage impacts the Fe–As coupling in stabilizing superconductivity in a different material.

High-quality single crystals of $KCa_2Fe_4As_4F_2$ were grown using the self-flux method, as reported previously.[7,39] The superconducting critical temperature at ambient pressure was confirmed to be 33.5 K, in good agreement with the results reported before.[7,10] High pressure was generated by a diamond anvil cell made of BeCu alloy with two opposing anvils. Diamond anvils with 300 μm culet (flat area of the diamond anvil) were employed for several independent measurements. In the high-pressure resistance and Hall measurements, the Van der Pauw method was used, following the technique described by Van der Pauw in Philips.[40] Platinum foil was used as the electrodes, a rhenium plate as the gasket, cubic boron nitride as the insulating material, and NaCl as the pressure medium. High-pressure x-ray diffraction (XRD) measurements were carried out at beamline 15U of the Shanghai Synchrotron Radiation Facility. A monochromatic x-ray beam with a wavelength of 0.6199 Å was employed for these measurements. The pressure for all measurements was determined using the ruby fluorescence method.[41]

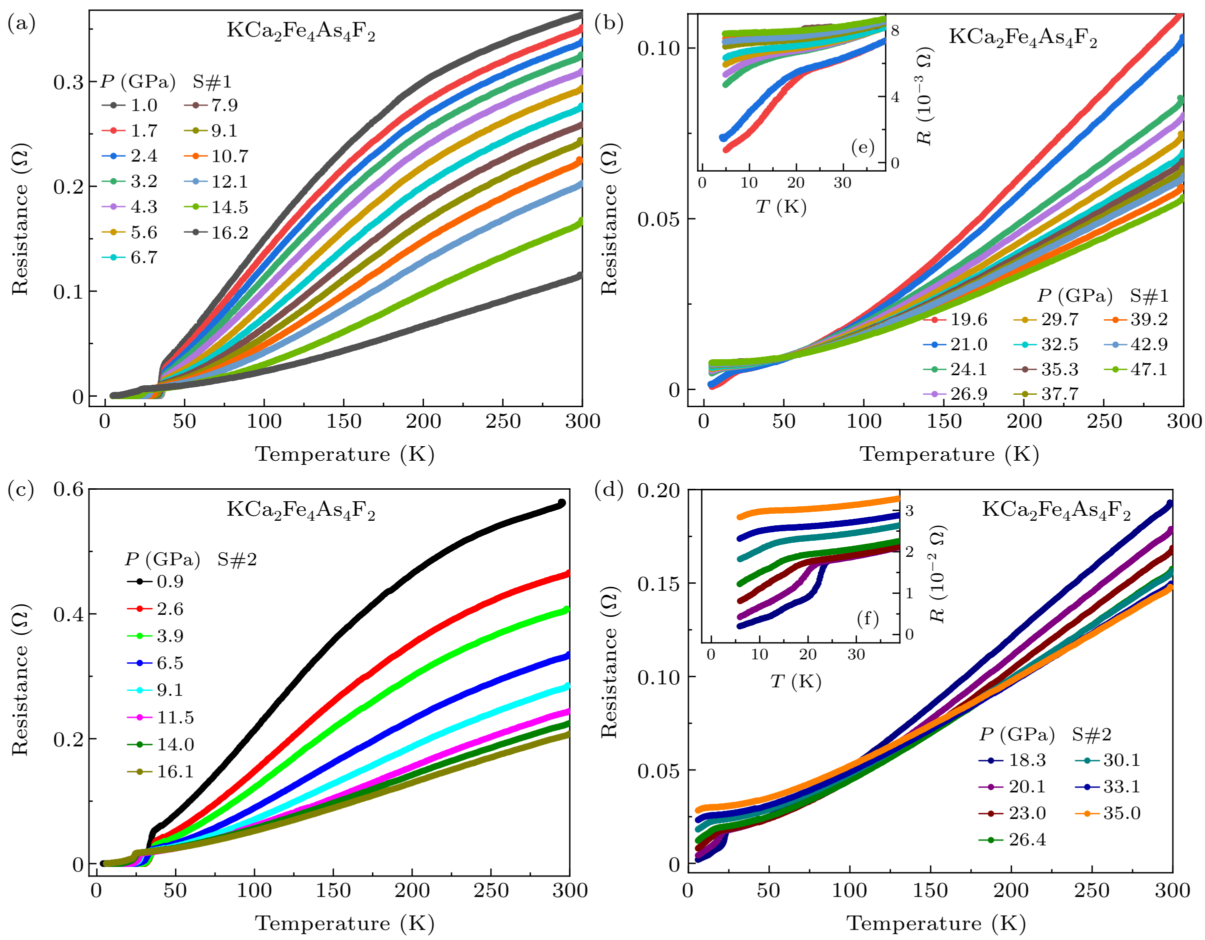

Fig. 1. Temperature dependence of resistance for the two $KCa_2Fe_4A_2$ samples under pressures ranging from 1.0 GPa–16.2 GPa (a) and 19.6 GPa–47.1 GPa (b) for S#1, 0.9 GPa–16.1 GPa (c) and 18.3 GPa–35.0 GPa (d) for S#2. The insets of panels (b) and (d) display low temperature resistances of the two samples.

First, we performed *in-situ* resistance measurements on the $KCa_2Fe_4As_4F_2$ samples in a diamond anvil cell. Figure 1 illustrates the variation in electrical resistance as a function of temperature for two samples (S#1 and S#2) at different pressures up to 47.1 GPa and 35.0 GPa, respectively. It is seen that the superconducting critical temperatures (T_c) of both samples increase initially. For S#1, T_c reaches a maximum of 37.5 K at around 2.4 GPa (Fig. 1(a)). Similarly, for S#2, T_c increases to 37.3 K at around 2.6 GPa (Fig. 1(c)). These observations align with the results reported in a previous study.[38] Upon further increasing the pressure, T_c exhibits a monotonic decrease (Figs. 1(b) and 1(d)) and is completely suppressed at pressure between 39.2 GPa and 42.9 GPa (Fig. 1(b)). Here we take the average value ($[39.2+42.9]/2 = 41.05$ GPa) as the critical pressure for the transition from superconducting to non-superconducting state.

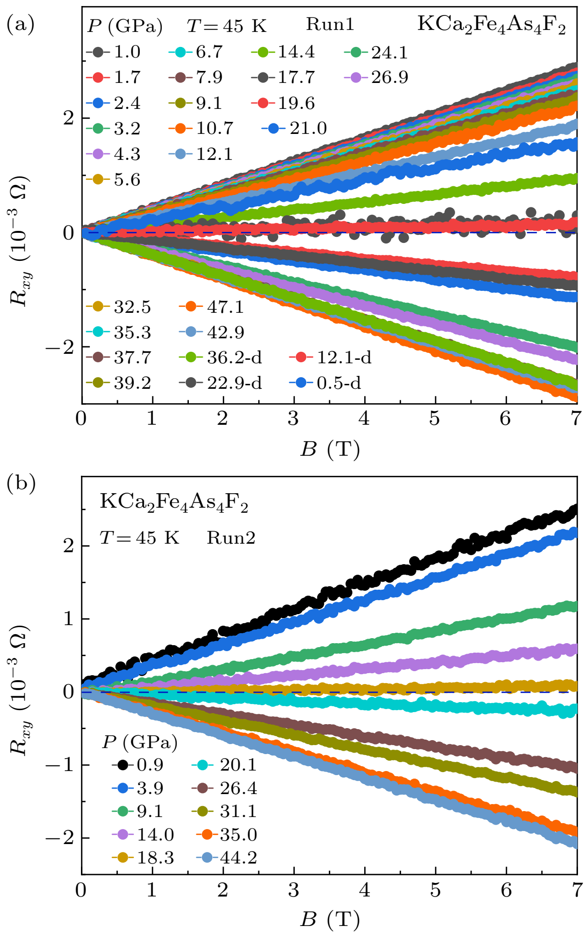

Fig. 2. Hall resistance (R_{xy}) as a function of magnetic field (B) for the two $KCa_2Fe_4As_4F_2$ samples at different pressures. In (a), the data labeled by 36.2-d, 22.9-d, 12.1-d and 0.5-d are the results obtained from the releasing pressure measurements.

To understand the pressure-induced change in T_c with normal state transport properties for the 12442 superconductor, high-pressure Hall resistance (R_{xy}) measurements were performed on the samples S#1 and S#2 through sweeping the magnetic field (B) applied perpendicular to the *ab* plane of those two samples from 0 T to 7 T at 45 K, as shown in Fig. 2. We found that the plot of R_{xy} versus B exhibits positive relationship below 17.7 GPa for S#1 and 18.3 GPa for S#2, indicating that hole carriers are dominated within the pressure range. However, $R_{xy}(B)$ changes its sign from positive to negative at about 17.7 GPa for S#1 and 18.3 GPa for S#2, revealing that the dominant carriers in this superconductor change from the hole-type into the electron-type.

To ensure a reasonable estimation, we consider the average value obtained from the two independent runs of measurements on the samples as the critical pressure ($P_c = [17.7+18.3]/2 = 18$ GPa). Based on these results, we propose that a significant Fermi surface reconstruction from a hole dominated to an electron dominated one occurs at about 18 GPa.

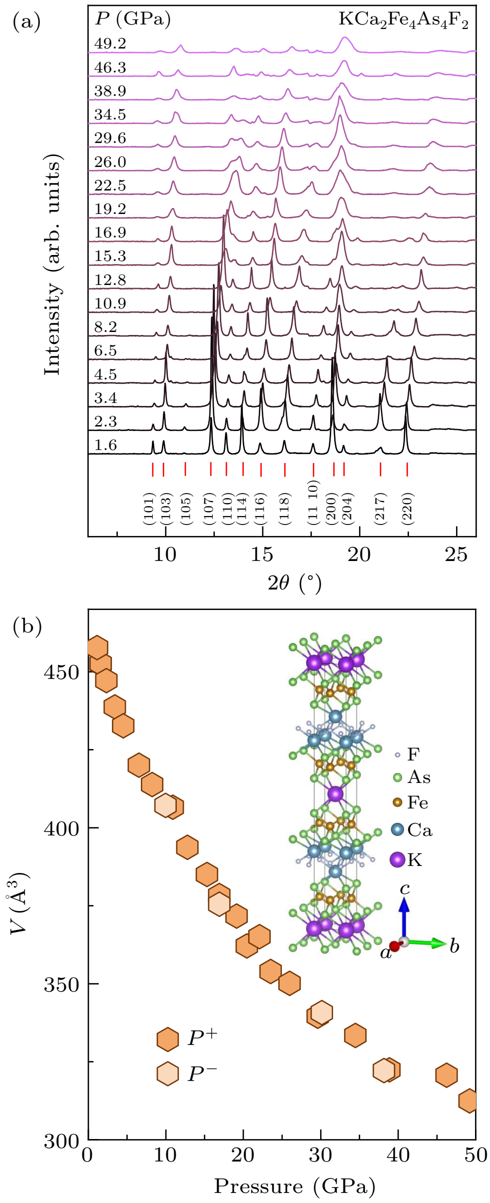

Fig. 3. XRD results of the $KCa_2Fe_4As_4F_2$ sample under different pressures. (a) XRD patterns collected at different pressures, showing no first-order structural phase transition up to 49.2 GPa. (b) The pressure dependence of volume (V). The inset illustrates the unique crystal structure of the 12442 superconductor.

Since the superconductivity usually has an intimate connection with the lattice structure, we conducted x-ray diffraction (XRD) measurements on the 12442 sample under pressures up to 49.2 GPa, with an aim to know how the changes in superconductivity and Hall resistance are accompanied by lattice shrinkage. Figure 3(a) displays the XRD results obtained from the 12442 sample at different pressures. We found that all the observed peaks can be indexed well by the tetragonal (T) phase in the $P4/mmm$ space group. Based on these results, we extracted the pressure dependence of lattice parameters (they will be discussed later) and volume (Fig. 3(b)). The gradual decrease in volume signifies the absence of any first-order phase transition within the pressure range up to 49.2 GPa.

We summarize our high-pressure experimental results obtained from the measurements on the 12442 superconductors in the pressure–T phase diagram (Fig. 4(a)). The diagram reveals three distinct regimes. In the low-pressure regime (below P_{c1}), the sample maintains its T phase, wherein the superconducting temperature T_c displays an increase initially from 33.5 K to 36.5 K at 2.4 GPa, followed by a steady decrease (Fig. 4(a)). Within this regime, we observed a reduction in both lattice parameters a and c (Fig. 4(b)) and the hole-type carriers are dominating (Fig. 4(c)).

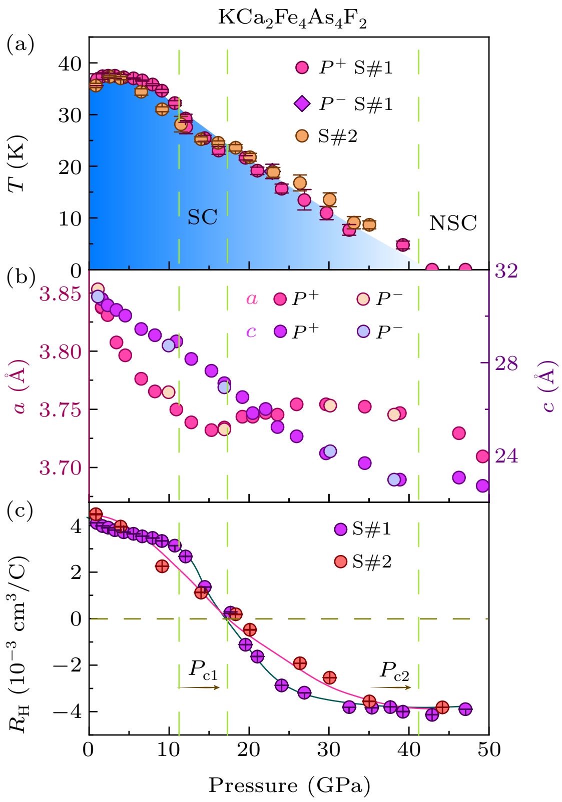

Fig. 4. Coevolution of superconductivity, crystal structure and Hall coefficient of $KCa_2Fe_4As_4F_2$ under pressures. (a) Pressure–temperature phase transition, revealing a pressure-induced change in T_c. (b) Pressure dependence of lattice parameters a and c, illustrating a pressure-induced half collapsed tetragonal phase starting at P_{c1} ($\sim$ 18 GPa) and full collapsed tetragonal phase at P_{c2} ($\sim$ 41 GPa). (c) Pressure dependence of Hall coefficient, showing the pressure-induced change from the hole-type carrier to the electron-type one at P_{c1}, and remaining unchanged at pressure above P_{c2}.

Intriguingly, at P_{c1}, we observe a notable expansion in the lattice parameter a (Fig. 4(b)). This phenomenon is reminiscent of what has been observed in the 1144 superconductors, where the lattice parameter a also undergoes a sudden increase while the volume continues to decrease. These observations suggest that the 12442 superconductor also undergoes an iso-structure transition from the tetragonal (T) phase to the collapsed tetragonal (cT) phase at P_{c1}. Interestingly, at this critical pressure, the Hall coefficient (R_H) undergoes a sign change from positive to negative, like what has been observed in the 1144 superconductors.[25–27,42–44] As the pressure is increased, we observe a gradual increase in the lattice parameter a (Fig. 4(b)), accompanied by a continued decrease in T_c (Fig. 4(a)). At P_{c2}, the sample loses its superconductivity ($T_c = 0$, see Fig. 4(a)), and the lattice parameter a begins to decline again (Fig. 4(b)), suggesting that pressure-induced collapse of the T phase is completed. This observation generally aligns with the behavior observed in the 1144 superconductor,[25,42,43] however the superconductivity in the latter is entirely suppressed upon a full collapse of the T phase. Upon further increasing the pressure beyond P_{c2}, we find that the lattice parameter a starts to decrease and R_H remains nearly unchanged (Fig. 4(c)).

Previous calculations have suggested that Fe–As coupling plays a pivotal role in developing or stabilizing superconductivity of iron pnictides.[11] Considering the unique structure of the 12442 superconductor, the pressure-induced transitions of the cT phase appear to promote the formation of the As–As bonding across the K layer, similar to what has been observed in the 1144 superconductors.[11,25,27,42] These bond formations inevitably weaken the Fe–As coupling, which significantly impacts the electronic structure and, consequently, the superconductivity. This experimental observation provides further evidence of the crucial role played by Fe–As coupling in stabilizing the superconductivity of iron pnictides. In addition, our results show that the pressure-induced coevolution of superconductivity and Hall coefficient with the lattice shrinkage is reversible, as presented in Figs. 3 and 4, which deserves further investigations.

In conclusion, we report the coevolution of superconductivity and Hall coefficient, along with the anisotropic lattice shrinkage, in compressed $KCa_2Fe_4As_4F_2$ up to 47.1 GPa. Our results clearly demonstrate that below the first critical pressure ($P_{c1} = 18$ GPa), superconducting transition temperature (T_c) displays an increase initially and then exhibits a monotonous decrease. Hall measurements find that the hole-type carriers are dominating in this pressure range. At around P_{c1}, we observed that the sample undergoes an iso-structure phase transition from its initial tetragonal (T) phase to a collapsed T (cT) phase, concurrently, the Hall coefficient undergoes a sign change from positive to negative. Upon increasing the pressure above P_{c1}, the superconducting transition temperature (T_c) exhibits a continuous decrease. At the second critical pressure ($P_{c2} = 41$ GPa), the sample completely loses its superconductivity, associated with the complete collapse of the T phase, while the Hall coefficient remains nearly unchanged. These results demonstrate that the pressure-induced anisotropic lattice collapse plays a crucial role in tuning the interplay among the multiple degrees of freedom in the superconducting system and, correspondingly, the stability of the superconductivity.

Acknowledgments

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