Nonmetallic Low-Temperature Normal State of $\text{K}_{0.7}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$

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The normal-state in-plane resistivity below the zero-field superconducting transition temperature T_c and the upper critical field $\mu_0 H_{c2}(T)$ was measured by suppressing superconductivity in pulsed magnetic fields for $\text{K}_{0.7}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$. The normal-state resistivity ρ_{ab} is found to increase logarithmically with decreasing temperature as $(T/T_c) \rightarrow 0$. Similar to granular metals, our results suggest that a superconductor–insulator transition below zero-field T_c may be induced in high magnetic fields. This is related to the intrinsic real-space phase-separated states common to all inhomogeneous superconductors.

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I. INTRODUCTION

The normal-state properties (including the pseudogap at the Fermi surface and the proximity of magnetism at the phase diagrams) of unconventional superconductors such as high- T_c copper oxides, iron-pnictides/chalcogenides, and heavy-fermions have received much attention [1]. The linear temperature dependence of the in-plane resistivity above T_c [2–4] is considered to be related to magnetic quantum criticality and antiferromagnetic fluctuations [5]. Since the normal (nonsuperconducting) states of interest appear under extremely high magnetic fields and at low temperatures ($T \rightarrow 0$), only a few studies have investigated the normal-state properties of high- T_c superconductors below T_c so far. In copper oxides, examples include underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_6$, where $\rho(T)$ diverges logarithmically below T_c and where a superconductor–insulator transition (SIT) is induced in pulsed magnetic fields [6–9]. This came as a surprise since the insulating state revealed by the magnetic-field-induced breakdown of superconductivity was not expected in a conventional metal. Studies of $(\text{Ba}, \text{K})\text{Fe}_2\text{As}_2$ and $\text{LaFeAsO}_{1-x}\text{F}_x$ resistivity suggested a slight upturn below zero-field T_c after the partial suppression of superconductivity up to a 61-T pulsed field [10,11]. In $\text{SmFeAsO}_{1-x}\text{F}_x$, magnetic-field-induced

logarithmic insulating behavior in the resistivity spans one decade in temperature above the zero-field T_c , which stems from the large magnetoresistance that extends to temperatures well above the log- T regime and is not linked to the superconducting state [12].

$\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ exhibits a high-temperature insulator-metal crossover at $T_s \sim 100\text{--}200$ K before becoming superconducting at $T_c = 30$ K [13–15]. Insulating ground states can also be found by tuning parameters such as Fe occupancy or isovalent S substitution on the Se atomic site [16,17]. As opposed to copper oxides, a dual description including both itinerant and localized carriers or magnetic moments may be required [18,19]. But similar to the copper oxides, it is imperative to probe the normal state below zero-field T_c . This is even more true because of the argued granularlike superconductivity arising in nanoscale superconducting grains separated by nonsuperconducting islands, with a macroscopic superconducting condensate being established via Josephson tunneling among the grains [20,21]. In the underdoped cuprates electronic granularity stems from the nature of the chemical bonds; hole-rich superconducting regions may form for low levels of hole doping even though the crystal structure is not granular [22]. In $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ crystals, the phase separation is an intrinsic feature of the crystal structure [23,24].

Here, we measured the normal-state in-plane resistivity below T_c for $\text{K}_{0.7}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ by suppressing superconductivity in pulsed magnetic fields. The normal-state resistivity ρ_{ab} is found to increase logarithmically as $(T/T_c) \rightarrow 0$. Similar to granular superconductors and underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_6$ [6–9], our results suggest for the first time that magnetic-field-driven

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SIT is induced in high magnetic fields in Fe-based superconductors.

II. EXPERIMENTAL

Single crystals of pure $K_xFe_{2-y}Se_2$ and nominally 10% Te-doped $K_xFe_{2-y}Se_2$ used in this study were grown and characterized as described previously [17]. The powder x-ray diffraction (XRD) spectra were taken with Cu $K\alpha$ radiation ($\lambda = 0.15418$ nm) by a Rigaku Miniflex x-ray machine, confirming phase purity. The cleaved surface image was determined in a JEOL JSM-6500 scanning electron microscope (SEM). The average stoichiometry was determined by energy-dispersive x-ray spectroscopy (EDX) in the same SEM. Four-probe resistivity was measured in pulsed magnetic fields up to 65 T and in Quantum Design PPMS-14 and PPMS-9. Contacts were attached to the ab plane of crystals in a glove box. Sample dimensions were measured by an optical microscope Nikon SMZ-800 with 10- μ m resolution. The electric current used in the pulsed-field measurement was always 2 mA, and the corresponding electric current density in the sample was about 1.39×10^4 A/m².

III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the diffraction patterns of both compounds, and all reflections can be indexed in the I4/mmm space group. The lattice parameters for $K_{0.70}Fe_{1.46}Se_{1.85}Te_{0.15}$ from the refinement are $a = b = 3.910(3)$ nm and $c = 14.268(6)$ nm, which are larger than for $K_xFe_{2-y}Se_2$ [$a = b = 3.881(1)$ nm and $c = 14.151(6)$ nm]. The expanded crystal lattice implies that Te enters the lattice. This is further confirmed by the EDX large-area elemental analysis, which shows the average stoichiometry $K_{0.70(6)}Fe_{1.46(3)}Se_{1.84(8)}Te_{0.15(2)}$ [Fig. 1(b)]. The SEM image [Fig. 1(c)] shows a typical phase-separation pattern. Compared to undoped $K_xFe_{2-y}Se_2$ [25–27], the pattern is inhomogeneous: Stripes can be up to 100 nm large, whereas their lower limit is below 50 nm. The local-point analysis on the background (dark) areas gives the stoichiometry $K_{0.78(4)}Fe_{1.70(3)}Se_{1.84(8)}Te_{0.15(2)}$, which is close to the 245 insulating phase [28]. About 7.5% Te substitution induces a $T_{c(onset)}$ decrease from about 30 K to about 29.5 K, and the metal-semiconductor transition temperature T_s decreases from about 130 K to about 100 K [Fig. 1(d)].

Near T_c , the upper critical field slopes, $d\mu_0 H_{c2}(T)/dT|_{T=T_c}$ in $K_xFe_{2-y}Se_2$ and other ternary or quaternary iron arsenide and selenide superconductors, are about 5 to 12 TK⁻¹ for $H||ab$ and 1 to 3 TK⁻¹ for $H||c$. Using the Werthamer-Helfand-Hohenberg (WHH) estimate [29] results in $\mu_0 H_{c2}(T)|_{T=0} = (125-275)$ T and (30–60) T for $H||ab$ and $H||c$, respectively [30–32]. The upper critical field for $H||c$ presents almost linear temperature dependence up to 60 T instead of the saturation predicted by WHH.

FIG. 1. A nominal 7.5% Te substitution of Se in $K_xFe_{2-y}Se_2$ induces a decrease of the superconducting transition temperature T_c and of the semiconductor-metal transition temperature T_s . (a) The powder x-ray diffraction patterns for Te-doped and pure $K_xFe_{2-y}Se_2$ crystals. The shift of the diffraction peaks toward lower angles shows the expanded lattice in Te-doped crystals. (b) The energy-dispersive x-ray spectroscopy of a typical $K_{0.70}Fe_{1.46}Se_{1.85}Te_{0.15}$ single crystal, which clearly shows the Te peak. (c) The SEM image of a typical single crystal showing stripe phases. (d) Temperature dependence of the resistivity for pure and Te-doped $K_xFe_{2-y}Se_2$. The inset shows the magnified part around the superconducting transition temperature. $T_{c(onset)}$ decreases from 30 to 29.5 K, and T_s decreases from 130 to 100 K.

This is ascribed to the multiband effect [30,31]. Although the upper critical field for $H||c$ is larger than the WHH estimate, it is smaller than $\mu_0 H_{c2}(T)$ for $H||ab$, which is explained well within the WHH theory [31,32]. For $K_{0.70}Fe_{1.46}Se_{1.85}Te_{0.15}$, we observe similar slopes of $\mu_0 H_{c2}(T)$ with $d\mu_0 H_{c2}(T)/dT|_{T=T_c} \sim 2.1(4)$ TK⁻¹ for $H||c$ [Figs. 2(a)–2(c)] and $d\mu_0 H_{c2}(T)/dT|_{T=T_c} \sim 4.9(3)$ TK⁻¹ for $H||ab$. However, in $K_{0.70}Fe_{1.46}Se_{1.85}Te_{0.15}$, at lower temperatures, $\mu_0 H_{c2}(T)$ is significantly reduced. A magnetic field of 61 T suppresses the superconductivity at 1.7 K for $H||c$, as opposed to $H||a$ [Fig. 3(a)]. This is ascribed to a change in the temperature dependence of $\mu_0 H_{c2}(T)$.

FIG. 2. Field dependence of the in-plane resistivity in a $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ single crystal with a magnetic field applied parallel to the c axis at fixed temperatures. (a) The resistivity data for temperatures between 22 and about 33 K up to 14 T. (b) Temperature dependence of $\mu_0 H_{c2}(T)$ near zero-field T_c for the Te-doped crystal with $H\parallel c$ using different criteria in the resistive transition shown in (a). (c) Resistivity data for temperatures between 1.75 and 33 K in pulsed fields up to 60 T.

Effects of Pauli spin paramagnetism and spin-orbit scattering can be included in the WHH theory for a single-band s -wave weak-coupling type-II superconductor in the dirty limit by adding the Maki parameters α and λ_{so} [29,33]. Then, $\mu_0 H_{c2}(T)$ is given by

$$\ln\left(\frac{1}{t}\right) = \left(\frac{1}{2} + \frac{i\lambda_{so}}{4\gamma}\right)\psi\left(\frac{1}{2} + \frac{h + \lambda/2 + i\gamma}{2t}\right) + \left(\frac{1}{2} - \frac{i\lambda_{so}}{4\gamma}\right)\psi\left(\frac{1}{2} + \frac{h + \lambda/2 - i\gamma}{2t}\right) - \psi\left(\frac{1}{2}\right), \quad (1)$$

where $\psi(x)$ is the digamma function, $\gamma = [(\alpha h)^2 - (\lambda_{so}/2)^2]^{1/2}$, and

$$h = \frac{4\mu_0 H_{c2}(T)}{\pi^2 T_c [-d\mu_0 H_{c2}(T)/dT]_{T=T_c}}. \quad (2)$$

In our data, we observe a convex curvature and a tendency for $\mu_0 H_{c2}(T)$ to saturate, with $H\parallel c$ at low temperatures [Fig. 3(b)], which can be well understood by the WHH theory with $\alpha = 0$ and $\lambda = 0$, i.e., with orbitally limited

FIG. 3. The upper critical fields in $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ follow the temperature dependence as predicted by the WHH theory. (a) Field dependence of the in-plane resistivity of $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ for pulsed magnetic fields applied perpendicular to the c axis. (b) Temperature dependence of the upper critical field $H_{c2}(T)$ (plotted as $(\mu_0 H_{c2}/T_c)[-d\mu_0 H_{c2}/dT]_{T_c}$) for magnetic fields parallel (circles) and perpendicular (balls) to the c axis, respectively. The solid lines are the fitting results using WHH theory with different parameters.

H_{c2} : $\mu_0 H_{c2}^*(0) = -0.693T_c(dH_{c2}/dT)_{T_c}$ [the blue curve in Fig. 3(b)]. Hence, the isovalent Te substitution suppresses the multiband character and promotes a single-band orbital-limited behavior.

The in-plane upper critical field is limited because of spin paramagnetism since $\alpha = 0.28$ and $\lambda_{so} = 0$ [red curve in Fig. 3(b)]. When compared to $\alpha_{H\parallel ab} = 5.6$ and $\alpha_{H\parallel ab} = 1.9$ in $\text{Ti}_{0.58}\text{Rb}_{0.42}\text{Fe}_{1.72}\text{Se}_2$ and $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, respectively [30,32], the spin-paramagnetic effect in the Te-doped system is very small ($\alpha < 1$), but the high anisotropy of $\mu_0 H_{c2}(T)$ for the different principal crystallographic directions remains, suggesting that an anisotropic Fermi surface is preserved. Open electron orbits along the c axis render an orbital-limited field unlikely, and spin-paramagnetic effects should play an important role for $H\parallel ab$. The paramagnetically limited field $\mu_0 H_{c2}^p(0)$ is given by $\mu_0 H_{c2}^p(0) = \mu_0 H_{c2}^*(0)/\sqrt{1 + \alpha^2}$ and $\alpha = \sqrt{2}H_{c2}^*(0)/H_p(0)$, where $\mu_0 H_p(0)$ is the zero-temperature Pauli limited field and the orbitally limited $\mu_0 H_{c2}^*$ given above [33]. This gives $\mu_0 H_{c2}^{p,ab}(0) = 100$ T, whereas $\mu_0 H_{c2}^{p,c}(0) = 45.7$ T. The zero-temperature coherence length $\xi(0)$ can be estimated using the Ginzburg-Landau formula $\mu_0 H_{c2}(0) = \Phi_0/2\pi\xi^2(0)$, where $\Phi_0 = 2.07 \times 10^{-15}$ Wb, resulting in $\xi(0)_{ab} = 2.7(1)$ nm and $\xi(0)_c = 1.8(3)$ nm, respectively.

The reduction of the Maki parameter α in $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$, when compared to $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, cannot be explained by disorder effects since for electronic systems with more disorder, one expects a larger α [34]. Therefore, another effect must compete and prevail over the

disorder induced by Te substitution. With strong-coupling corrections for electron-boson and electron-electron interactions, $\mu_0 H_p(0) = 1.86(1 + \lambda)^\varepsilon \eta_\Delta \eta_{ib}(1 - I)$, where strong-coupling intraband corrections for the gap are described by η_Δ , I is the Stoner factor $I = N(E_F)J$, $N(E_F)$ is the electronic density of states (DOS) per spin at the Fermi level, E_F , J is an effective exchange integral, η_{ib} is introduced to phenomenologically describe the effect of the gap anisotropy, λ is the electron-boson coupling constant, and $\varepsilon = 0.5$ or 1 [35]. The spin-paramagnetic effect can be less pronounced when the Stoner factor becomes small, which can be achieved through suppressing either the DOS at the Fermi level or the effective exchange integral J [35]. The reduction of both the multiband character in $H_{c2}(T)$ and the spin paramagnetism, when compared to pure $K_x\text{Fe}_{2-y}\text{Se}_2$, implies that electronic states, removed by Te substitution, carry substantial partial density of states at the Fermi level, i.e., that Te substitution reduces the occupancy of heavily renormalized d_{xy} orbitals [36].

In $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$, a 61-T magnetic field parallel to the c axis completely suppresses superconductivity above 1.75 K [Figs. 2(c) and 3(b)], providing the opportunity to study the normal-state properties as $T \rightarrow 0$. The application of 61 T leads to a negative $d\rho/dT$ resembling insulating rather than metallic behavior [Fig. 4(a)]. It is apparent that a SIT occurs right below T_c (filled balls) after the superconductivity is completely suppressed above H_{c2} . At 50 T, the resistivity also shows SIT and is nearly identical to that at a 61-T field until superconductivity sets in below about 15 K. We first note that the insulating behavior in high magnetic fields appears only below the zero-field T_c . It is also instructive to note that above the zero-field T_c , the field dependence of the resistivity up to 61 T is minute for $H\parallel c$, in contrast to $\text{SmFeAsO}_{1-x}\text{F}_x$ [12]. The temperature-magnetic-field phase diagram for $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ is shown in Fig. 4(b).

A SIT was first reported in ultrathin, disordered and granular type-I superconductors [37,38]. Then, a SIT with an insulating resistivity, increasing logarithmically below T_c , was reported in high- T_c cuprate superconductors utilizing pulsed magnetic fields [6–8]. Such a temperature dependence is different from variable range hopping (VRH) and thermally activated transport observed in semiconductors. Kondo-type magnetic scattering is also unlikely since the spin-flip scattering should be suppressed at 61 T. Evidence for an intrinsic electronic phase separation in the cuprate superconductors [39] suggests that the logarithmic increase in the resistivity and the SIT originates from an inhomogeneous state, presumably similar to granular Nb [38,40].

$\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ features a highly defective crystal structure, where the magnetic insulating regions are spatially intrinsically separated and coexist with superconducting regions on a length scale of 100 nm to 1 μm [25–27]. However, as opposed to two-dimensional alternate stacking of insulating

FIG. 4. (a) Temperature dependence of the in-plane resistivity in several DC (open symbols) and pulsed (filled symbols) magnetic fields for temperatures between 1.75 and 100 K. Pulsed-field data (filled symbols) near T_c are consistent with results in DC and zero fields (open symbols). The two half-filled symbols for 61 T at 1.75 and 4 K denote the extrapolated values of the resistivity after complete suppression of the superconductivity. The red and green dashed lines are the linear fitting result for the 61 T data and 50 T data when using the logarithmic scale, respectively. Inset: Resistivity of the mixed state in a 50-T field. The blue dotted line is an exponential fitting result (see text). (b) The schematic temperature-field phase diagram of $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$.

and (super)conducting layers, the stripe-type phase separation in $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ is more three dimensional [21–23]. The conductivity in such inhomogeneous samples must include contributions from the insulating (σ_i) and metallic (σ_m) regions, i.e., $\sigma = \sigma_m + \sigma_i$. When $T < T_c$, σ_m goes to infinity because of superconductivity, and the insulating part of the sample is short circuited ($\sigma = \sigma_m = \infty$). But even around T_c , and, in particular, in the normal state at high magnetic fields when $T \rightarrow 0$, $\sigma \sim \sigma_m$ holds as well since insulating regions have a nearly 2 orders of magnitude higher resistivity [41]. This is consistent with the observation that insulating regions do not contribute to the spectral weight in angular-resolved photoemission data close to E_F [36].

Therefore, superconductivity in $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$ is similar to that in granular superconductors, namely, a three-dimensional array of superconducting grains in an insulating matrix [38,40]. Strong magnetic fields suppress superconductivity in each grain. The possible scenarios explaining a SIT include Anderson localization [42] and the bosonic scenario where the gap is driven to zero by phase fluctuations [43,44]. The observed logarithmic divergence of the resistivity in the nonmetallic normal state of $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ [Fig. 4(a)] does not follow the VRH conduction that is observed for Anderson localization. In the bosonic scenario, the Cooper pairs are

localized in granules (similar to defects). For $H > H_{c2}$, virtual Cooper pairs may form, leading to a reduction of the DOS. When $T \rightarrow 0$, the pairs cannot travel between grains, and the resistivity becomes much larger than for a normal metal without electron-electron interactions [43,44]. In contrast to the spin-paramagnetic critical field, the orbital critical field depends on the grain size as $H_{c2}^o \sim \phi_0/R\xi$, where R is the average grain radius and ξ is the coherence length. Since the upper critical field in $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ is about 55(3) T and the grain coherence length is 2.7(1) nm, we estimate the lower limit of the stripe grain size to be $R = 21(2)$ nm. Near the SIT on the superconducting side, it is expected that $\rho = \rho_0 \exp(T/T_0)$ [40] in high fields because of the destruction of fluctuating quasilocalized Cooper pairs and a concomitant increase of the number of quasiparticles at the Fermi level. Our 50-T resistivity data in the mixed state can be described well by this formula [the blue line in the inset of Fig. 4(a)]. Different heat treatment and quenching can bring about changes in the normal-state resistivities, arrangement, and connectivity of the superconducting grains [28]. This can influence the tunneling conductance in the granular array, and therefore, it would be of great interest to study superconducting fluctuations in such samples in high magnetic fields in the normal state below T_c for further comparison with theory [38,40].

Even though the logarithmic temperature dependence of the resistivity in copper oxides is consistent with the theory of granular superconductivity if electrons in different CuO planes are incoherent in the low-temperature limit [45,46], this is still controversial since the SIT in copper oxides occurs on the metallic side of the Mott limit ($k_F l > 1$, where k_F is the Fermi wave vector and l is the mean free path) [47]. Approaches offered to explain insulating states in copper-oxide superconductors postulate spin-charge separation [48], invoke a bipolaron theory of strongly correlated Mott-Hubbard insulators [49], or propose Mott insulating states in antiferromagnetic vortices within the framework of $\text{SO}(5)$ theory of antiferromagnetism and superconductivity [50]. In the light of increasing evidence for insulating states in the different kinds of high- T_c superconductors in pulsed magnetic fields, more theoretical work may be needed to determine which of these approaches is compatible with real-space and/or electronic phase separation in materials of different structure and bonding types.

IV. CONCLUSION

In summary, the normal-state in-plane resistivity and the upper critical fields of $\text{K}_{0.70}\text{Fe}_{1.46}\text{Se}_{1.85}\text{Te}_{0.15}$ are measured by suppressing superconductivity in pulsed magnetic fields. The temperature dependence of $\mu_0 H_{c2}(T)$ can be well described using the WHH theory, in contrast to $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, where the linear temperature dependence signals multiband effects. After suppressing superconductivity, the normal-state resistivity ρ_{ab} is found to be

insulating and to diverge logarithmically as $(T/T_c) \rightarrow 0$. Similar to the SIT in granular superconductors, our results suggest that the mechanism for the SIT in high magnetic fields is related to intrinsic phase-separated states in these kinds of materials.

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