Evolution of correlation strength in $K_x Fe_{2-y} Se_2$ superconductor doped with S

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We report the evolution of thermal transport properties of $K_x Fe_{2-y} Se_2$ with sulfur substitution at Se sites. Sulfur doping suppresses the superconducting T_c . The Seebeck coefficient of all crystals in the low-temperature range can be described very well by a diffusive thermoelectric response model. The zero-temperature extrapolated value of the Seebeck coefficient divided by temperature, S/T, gradually decreases from $-0.48 \,\mu\text{V/K}^2$ to a very small value, $\sim 0.03 \,\mu\text{V/K}^2$, where T_c is completely suppressed. The normal-state electron Sommerfeld term (γ_n) of specific heat also decreases with the increase in sulfur content. The decrease of S/T and γ_n reflects a suppression of the density of states at the Fermi energy, or a change in the Fermi surface that would induce the suppression of correlation strength. Our results imply little relevance of strong electron correlations to superconductivity.

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I. INTRODUCTION

Superconductivity in pure and F-doped LaFeAsO with T_c up to 26 K has opened a frontier in the investigation of superconducting materials and mechanisms.¹⁻³ After an intensive study, superconductivity was discovered in several different types of iron-based materials, including ROFePn (R = rare earth; Pn = P or As, 1111-type), $^{4-8}$ doped AFe_2As_2 (122-type, A = Ba, Sr, Ca), $^{9-11}$ Fe_2As -type AFeAs (111-type, A = Li or Na), 12,13 as well as anti-PbO-type Fe(Se,Te)(11-type). 14,15 All have a structure similar to the common FeAs-layer units. Most undoped compounds are stripelike antiferromagnetic [spin-density wave (SDW)] metals, and the magnetic ordering occurs in the vicinity of the structural phase transition from a tetragonal to an orthorhombic unit cell. Experimental and theoretical studies suggest that the high- T_c superconductivity in iron-based superconductors is influenced by their proximity to SDW phase transition. The doping brings along charges that suppress the SDW ordering. It was suggested that the superconductivity may be established via interpocket scattering of electrons between the hole pockets and electron pockets, leading to the s^{\pm} pairing. ^{16–18}

The electron correlation strength in parent materials is therefore one of the central problems in iron-based superconductors. The absence of strong correlations was noted within a tight-binding model and density functional calculations. ¹⁹ Moderate electron correlations in arsenic systems were also supported by a small observed ratio between the band-theoretical and experimental kinetic energy. ^{20,21} Theoretical studies gave an onsite Coulomb repulsion U < 2 eV versus an Fe conduction bandwidth $W \sim 4$ eV (i.e. $\frac{U}{W} < 0.5$), in 1111 and 122 systems. ^{22,23} The other line of study indicated that some iron-based superconductors have electron correlations comparable in strength to the cuprates. ^{24–27} The bad metal conductivity in the parent compounds was explained by the proximity to a Mott- or correlation-induced insulating state and strong electron correlations. ²⁴

Superconductivity with relatively high $T_c \sim 30$ K was recently reported in a series of iron-based superconductors, $A_x \text{Fe}_2 \text{Se}_2$ (A = K, Rb, Cs, Tl). These compounds are purely electron-doped and only electron pockets were observed in angle-resolved photoemission experiments. Just like other iron-based superconductors, the superconductivity in $A_x \text{Fe}_2 \text{Se}_2$ is sensitive to the Pn doping and anion

height between Fe and Pn layers. Sulfur doping at Se sites suppresses the superconductivity and induces a spin-glass narrow-band-gap semiconductor ground state for complete S substitution. The strength of electron correlations in $K_x Fe_{2-y} Se_2$ is also highly debated. Some studies indicated the superconductivity is in proximity to a Mott antiferromagnetic insulator, implying strong electron correlations. Others argued that the parent compounds are simple band insulators since density functional theory calculation could reproduce the experimental antiferromagnetic ground state with the iron vacancy order. Calculation of the local density approximation incorporating local interaction found that the band gap is nearly independent of the value of the Coulomb repulsion U, indicating the irrelevance of the Mott physics to gap formation in neighboring nonmetallic states.

Measurement of thermoelectric properties is an efficient method to characterize the correlation strength in superconductors, as well as the nature and sign of carriers. 25,42,43 In this work we clarify the correlation strength in S-doped $K_xFe_{2-y}Se_2$. $K_xFe_{2-y}S_2$ exhibits a large Seebeck coefficient at a high temperature range, which is attributed to the thermally activated carriers over the narrow band gap. In $K_xFe_{2-y}Se_{2-z}S_z$ ($0 \le z \le 2$) single-crystal alloys, the Seebeck coefficient and the electron Sommerfeld term in specific heat are suppressed as S enters the $K_xFe_{2-y}Se_2$ superconducting lattice. However, superconductivity survives with relatively high T_c even if electron correlations are considerably reduced, implying little relevance of the strong electron correlation to T_c .

II. EXPERIMENT

Single crystals of $K_x Fe_{2-y} Se_{2-z} S_z$ were grown from nominal composition K:Fe:Se:S=0.8:2:2 – z:z with different S content, as described elsewhere.^{36,37} The elemental analysis was performed using energy-dispersive x-ray spectroscopy in a JEOL JSM-6500 scanning electron microscope and in what follows we present the measured stoichiometry values (Table I). Electrical and thermal transport measurements were conducted in a Quantum Design PPMS-9. The crystal was cleaved to a rectangular shape with dimension $5 \times 2 \text{ mm}^2$ in the ab plane and 0.3 mm thickness along the c axis. Thermoelectric power and thermal conductivity were measured using a one-heater, two-thermometer setup, which allowed us to

Parameter	$K_{0.64}Fe_{1.44}Se_{2} \\$	$K_{0.73}Fe_{1.44}Se_{1.68}S_{0.32}$	$K_{0.70}Fe_{1.55}Se_{1.01}S_{0.99} \\$	$K_{0.76}Fe_{1.61}Se_{0.96}S_{1.04}$
$\frac{S}{T} (\mu V/K^2)$	-0.48(3)	-0.38(8)	-0.11(5)	-0.13(7)
$\gamma \text{ (mJ/mol } K^2)$	6.0(5)	3.5(7)	2.8(7)	1.5(4)
q	0.13	0.09	0.12	0.15
$T_c(K)$	31.0	31.4	21.4	16.4
$T_F(\mathbf{K})$	880	1110	3860	3270
$\frac{T_c}{T_F}$	0.04	0.028	0.005	0.005
$m^* (m_e)$	3.4	2.7	1.1	1.3

TABLE I. Set of derived parameters for superconducting $K_x Fe_{2-y} Se_{2-z} S_z$ crystals.

determine all transport properties of the crystal with the steady-state method. The heat and electrical current were transported within the ab plane of the crystal oriented by Laue camera, with the magnetic field along the c axis and perpendicular to the heat and electrical current. Silver paint contacts were made directly on the crystal surface, providing both good thermal contact and low electrical contact resistance. Since air exposure exceeding 1 h results in surface oxidization, the duration of the exposure to air of crystals was less than 20 min. The relative error in our measurement for both κ and S was less than 5% based on a Ni standard measured under identical conditions.

III. RESULTS AND DISCUSSIONS

Figure 1(a) presents temperature dependence of thermal conductivity for $K_x Fe_{2-y} Se_{2-z} S_z$ in zero magnetic field from 2 to 300 K. Thermal conductivity for all crystals exhibits a peak between 15 and 30 K [Fig. 1(b)]. The peak position moves to higher temperature with the increase of sulfur doping (red) arrows in [Fig. 1(b)]. It changes from \sim 17 K in undoped crystal to \sim 25 K in $K_x Fe_{2-y} S_2$ and is considered to entirely originate from phonon contribution. This shift is due to the lattice contraction corresponding to the smaller radius of the sulfur ion.

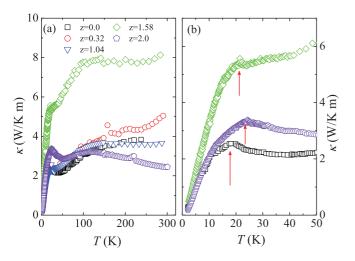


FIG. 1. (Color online) (a) Temperature dependence of thermal conductivity for $K_x Fe_{2-y} Se_{2-z} S_z$ in zero magnetic field from 2 to 300 K. (b) Low-temperature thermal conductivity with phonon-related peak indicated by (red) arrows.

With the increase of S concentration, superconducting T_c is suppressed and ultimately vanishes at z = 1.58 [Fig. 2(a)]. The suppression of T_c is confirmed in the Seebeck coefficient S=0 temperature [Fig. 2(b)] since Cooper pairs carry no entropy in the superconducting state. With increase in sulfur doping, the magnitude of the Seebeck coefficient decreases significantly for superconducting samples. The Seebeck coefficient for a crystal with z = 1.58 (which does not exhibit a superconducting transition above 1.9 K) is nearly zero in the whole temperature range (the value is \sim -0.75 μ V/K at 200 K and \sim 0.06 μ V/K at 2 K). For the narrow-band-gap semiconductor $K_{0.88}$ Fe_{1.63} S_2 , ρ and S are beyond the detection limit of our instrument, as shown in Fig. 2(b). The observable Seebeck coefficient appears at ~ 100 K and is due to the thermal excitation of carriers over the band gap. With further increase of temperature, the number of the thermally excited carriers becomes larger and the Seebeck coefficient increases up to $\sim 180 \ \mu V/K^2$ at 300 K. For all crystals, there are no evident peaks in the Seebeck curves between 2 and 300 K, indicating that there is no significant Fermi surface nesting in this temperature range. 25,43,44

The Seebeck coefficient in a material is the sum of three different contributions: the diffusion term $S_{\rm diff}$, the spin-dependent scattering term, and the phonon-drag term $S_{\rm drag}$ due to electron-phonon coupling. ^{43,45} Thermoelectric power (TEP) in our sample above T_c is independent of magnetic

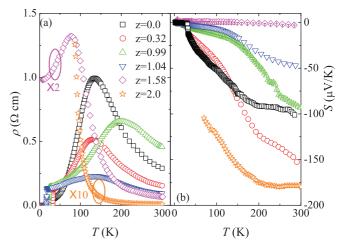


FIG. 2. (Color online) Temperature dependence of (a) resistivity and (b) Seebeck coefficient for $K_x Fe_{2-y} Se_{2-z} S_z$ (z=0.0, 0.32, 0.99, 1.04, and 1.58) under zero magnetic field.

^aValue obtained from Ref. 46.

field, which excludes the spin-dependent mechanism. The contribution of the phonon-drag term gives $\sim T^3$ dependence for $T \ll \Theta_D$, $\sim 1/T$ for $T \geqslant \Theta_D$ (where Θ_D is the Debye temperature), and a peak structure for $\sim \frac{\Theta_D}{5}$. ⁴⁵ The absence of the peak structure in our TEP results suggests a negligible contribution of the phonon-drag effect to S(T) since Θ_D for crystals with z=0 and z=2.0 are 260 K and 289 K, respectively. ^{37,44} At low temperature, the diffusive Seebeck response of a Fermi liquid dominates and is expected to be linear in T in the zero-temperature limit, with a magnitude proportional to the strength of electron correlations. ⁴² This is similar to the T linear electronic specific heat, $C_e/T=\gamma$. In a one-band system both can be described by

$$S/T = \pm \frac{\pi^2}{2} \frac{k_B}{e} \frac{1}{T_F} = \pm \frac{\pi^2}{3} \frac{k_B^2}{e} \frac{N(\epsilon_F)}{n},$$
 (1)

$$\gamma = \frac{\pi^2}{2} k_B \frac{n}{T_F} = \frac{\pi^2}{3} k_B^2 N(\epsilon_F), \tag{2}$$

where k_B is Boltzmann's constant, e is the electron charge, n is the carrier density, and T_F is the Fermi temperature, which is related to the Fermi energy ϵ_F and the density of states $N(\epsilon_F)$ as $N(\epsilon_F) = \frac{3n}{2\epsilon_F} = \frac{3n}{k_B T_F}$. ⁴² In a multiband system, this gives the upper limit of the Fermi temperature of the dominant band. The Seebeck coefficients of all crystals fit this formula very well in the low-temperature range. Figure 3(a) shows the relationship between the Seebeck coefficient divided by temperature (S/T) in $K_x Fe_{2-y} Se_{2-z} S_z$ with different S content under 0 T and 9 T magnetic fields, respectively. For superconducting crystals, the Seebeck coefficient in the normal state is independent of magnetic field and exhibits a linear relationship with temperature in the low-temperature range [Fig. 3(a)]. The zero-temperature extrapolated values of S/T for different crystals are shown in Fig. 3(b) and Table I. With sulfur doping, $\frac{S}{T}$ is suppressed from $-0.48 \ \mu V/K^2$ to a very small value of $\sim 0.03 \ \mu V/K^2$ for crystals without superconducting transition. A similar trend was observed in the suppression of superconducting T_c [Fig. 3(b)].

Our crystals do not exhibit a specific heat anomaly at the superconducting transition, similar to previous reports and possibly due to the very small superconducting contribution and the nodeless gap. 46 Yet the magnetic field-dependent specific heat can yield important information about the Fermi surface. Figures 4(a) and 4(b) show the specific heat data plotted as $\frac{C}{T}$ versus T^2 in the low-temperature region in different magnetic fields for crystals with z = 0.32 and z = 0.99 with upper critical field $H_{c2} \sim 45$ T and $H_{c2} \sim 13$ T (for a field applied along the c axis of the crystals), respectively.⁴⁷ The magnetic field gradually enhances the specific heat, indicating the buildup of the quasiparticle density of states. From linear fitting to C/T versus T^2 [solid lines in Figs. 4(a) and 4(b)], we obtained the linear dependence of the Sommerfeld coefficient on the magnetic field [Fig. 4(c)]. This is consistent with the results for $K_x Fe_{2-y} Se_2$ and the nodeless gap. 46 The slope of the line in Fig. 4(b) is $\sim 0.06(5)$ and $\sim 0.22(3)$ mJ/mol K² for two crystals with z = 0.32 and z = 0.99, respectively. We estimate the value of the normal-state electron specific heat coefficient γ_n to be 3.5 mJ/mol K² using an upper critical field $H_{c2}(0) \sim 45 \text{ T for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } \gamma_n = 2.8(7) \text{ mJ/mol K}^2 \text{ for } z = 0.32, \text{ and } z =$ a crystal with z = 0.99 using $H_{c2}(0) \sim 13$ T, respectively.⁴⁷

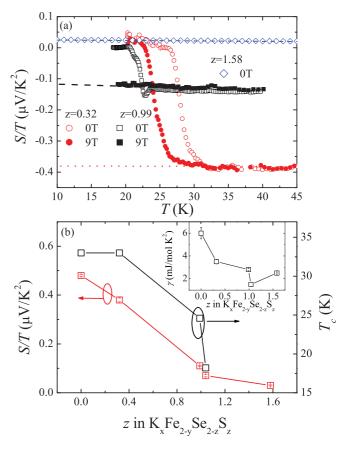


FIG. 3. (Color online) (a) Temperature dependence of the Seebeck coefficient divided by T, $\frac{s}{T}$, for $K_x Fe_{2-y}Se_{2-z}S_z$ with z=0.32, 0.99, and 1.58 under 0 T (open symbols) and 9 T (solid symbols), respectively. The dashed lines are the linear fitting results within the high-temperature range as described in text. (b) The relationship between the zero-temperature extrapolated value of $\frac{s}{T}$ (open circle) and superconducting T_c (open square) to S concentration z in $K_x Fe_{2-y}Se_{2-z}S_z$. The inset shows the relationship between the Sommerfeld coefficient and S concentration z in $K_x Fe_{2-y}Se_{2-z}S_z$. For superconducting crystals, the Sommerfeld coefficients in the normal state induced by magnetic field were used.

These values are smaller than the value [\sim 6.0(5) mJ/mol K²] in the K_xFe_{2-y}Se₂ system as shown in Table I and in the inset of Fig. 3(b).⁴⁶ Application of a 9 T magnetic field parallel to the *c* axis completely suppresses the superconductivity in the crystal with z=1.04 and we obtain $\gamma_n \sim 1.5$ mJ/mol K² directly from the linear fit of the low-temperature C/T versus T^2 in 9 T [Fig. 4(d)]. For a crystal with z=1.58, we extract the Sommerfeld coefficient from zero-field specific heat data [Fig. 4(d)]. The results are shown in Table I and in the inset of Fig. 3(b). With the increase in sulfur content the electronic Sommerfeld coefficient in the normal state is gradually suppressed.

According to Eqs. (1) and (2), S/T and the electron Sommerfeld term in specific heat are related to the carrier density and the density of states at the Fermi energy. Since sulfur has an identical electronic configuration to selenium, there should be no change in the carrier concentration with sulfur doping because the elemental analysis is consistent with full occupancy of S (Se) sites.³⁶ The absolute value of the

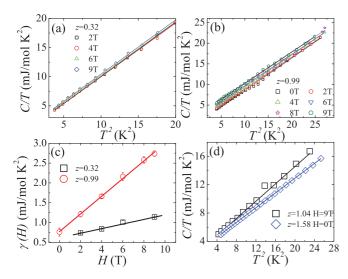


FIG. 4. (Color online) Low-temperature specific heat in different magnetic fields up to 9 T for crystals with (a) z=0.32 and (b) z=0.99. The lines are the linear fitting results. (c) Field dependence of the Sommerfeld coefficient $\gamma(H)$ for crystal with z=0.32 (squares) and z=0.99 (circles). (d) Low-temperature specific heat data for crystal with z=1.04 in 9 T, and for crystal with z=1.58 in 0 T field, respectively. The lines are linear fitting results.

dimensionless ratio of the Seebeck coefficient to specific heat, $q = \frac{N_{Av}eS}{T\gamma_n}$, with N_{Av} the Avogadro number, gives the carrier density. From the values of S/T and γ_n obtained previously, we derived q values for four superconducting crystals (Table I). The q values do not exhibit significant change. Therefore, the suppression of S/T and γ_n reflects a suppression of the density of states at the Fermi level or a change in the Fermi surface.

The ratio of the superconducting transition temperature T_c to the Fermi temperature T_F gives information about the correlation strength in superconductors. The ratio $\frac{T_c}{T_F} \sim 0.04$ for $K_{0.64}Fe_{1.44}Se_2$ implies a weakly correlated superconductor. With increase in sulfur content, the value of $\frac{T_c}{T_F}$ decreases as shown in Table I. This implies a suppression of electron correlation strength as the system is tuned toward semiconducting states. The effective mass m^* , derived from $k_B T_F = \frac{\hbar^2 k_F^2}{2m^*}$, is also suppressed with the increase in S content (Table I),

consistent with the decrease of correlation strength with S doping. This implies that the neighboring nonmetallic states, such as in $K_{0.88}Fe_{1.63}S_2$, do not originate from strong correlations as in a Mott insulator. Moreover, with the S increase from 0 to 0.8, $\frac{T_c}{T_F}$ decreases significantly (Table I), but the superconducting T_c survives and is relatively high (~20 K). This indicates that the electron correlations are not strong and do not play a primary role in the superconductivity. Recent first-principles calculation points out that the gap in the insulating states in proximity to $K_xFe_{2-y}Se_2$ does not depend on the Coulomb repulsive U, implying irrelevance of the Mott-insulator scenario.⁴¹ Our results are consistent with this theoretical study.

IV. CONCLUSION

In summary, we studied the evolution of thermal transport and thermodynamic properties of $K_x \text{Fe}_{2-y} \text{Se}_{2-z} S_z$ ($0 \le z \le$ 2). The zero-temperature extrapolated value of the Seebeck coefficient S/T is gradually suppressed and then undergoes a sharp decrease at z = 0.99 to a very small value $[\sim 0.03(2) \ \mu V/K^2]$ for crystals with more sulfur content. The electron Sommerfeld term (γ_n) in the specific heat also decreases with increase in sulfur content. The suppression of S/T and γ_n reflects a suppression of the density of states at the Fermi level or a change of Fermi surface. The ratio $\frac{T_c}{T_E}$ is also suppressed by sulfur doping, indicating the suppression of electron correlations. The superconductivity survives with relatively high T_c even when electron correlations are greatly reduced. This implies that this superconducting system does not lie in the proximity of a Mott insulator and that the electron correlations do not play a primary role in superconductivity.

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