Pressure dependence of the thermoelectric power of the iron-based high-$T_c$ superconductor SmFeAsO$_{0.85}$

N Kang$^{1,3}$, P Auban-Senzier$^1$, C R Pasquier$^1$, Z A Ren$^2$, J Yang$^2$, G C Che$^2$ and Z X Zhao$^2$

$^1$ Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France
$^2$ National Laboratory for Superconductivity, Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100190, People’s Republic of China
E-mail: kang@lps.u-psud.fr

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Abstract. We have measured the thermoelectric power $S$ of the iron-based superconductor SmFeAsO$_{0.85}$ as a function of temperature at various pressures up to 1.1 GPa. The magnitude of thermoelectric power increases with decreasing temperature and exhibits a maximum at a characteristic temperature $T^*$ ($\sim 110$ K at ambient pressure), whereas the temperature dependence of the resistance shows metallic behavior. The superconducting transition temperature $T_c$ and $T^*$ decrease monotonically with pressure. We discuss our results in terms of the effects of Fermi-surface nesting and orbit degeneracy in the new iron-oxypnictides.

Contents

1. Introduction ........................................ 2
2. Experimental ....................................... 2
3. Results and discussion .......................... 3
4. Conclusion ......................................... 6
Acknowledgments .................................. 7
References ........................................... 7

$^3$ Author to whom any correspondence should be addressed.
1. Introduction

Soon after the discovery of the superconductor LaFeAsO$_{0.89}$F$_{0.11}$ with $T_c \sim 26$ K [1], a new family of iron arsenide superconductors has been the subject of intensive research [2]–[9]. An important similarity to cuprate superconductors is that the iron-based superconductors ReFeAsO (Re = Sm, Pr, Ce and La) have also a layered crystal structure, where FeAs and ReO planes are alternately stacked along the $c$-axis. Direct studies of the Fermi surface in angle-resolved photoemission measurements have indicated the electron and hole pockets on the Fermi surfaces [10]. Therefore, the nesting of Fermi surface and its role in this system are of great interest. A knowledge of the behavior of electrons in the normal state seems to be essential for understanding the nature of transport processes. Thermolectric power (TEP) measurements under pressure are useful for investigating the superconducting behavior as well as transport properties in the normal state. Pressure experiment has drawn much attention due to the possibility of obtaining higher superconducting transition temperature $T_c$ [11]. It is important to determine how the pressure changes the band structure parameters as well as the different competing states and how these changes are related to the emergence of the superconducting state.

When a temperature gradient $\Delta T$ is applied to the sample, the charge would build up along the gradient. The thermopower is given by $S = \lim_{\Delta T \to 0} \Delta V/\Delta T$, where $\Delta V$ and $\Delta T$ are the potential and temperature differences across the sample, respectively. As a transport measurement, TEP is a powerful tool to provide important information related to the change of the electronic band structure at the Fermi energy, phase transitions and electron–phonon coupling. Moreover, the TEP can be used to determine the entropy flux accompanying charge transport. Recent experiments on LaFeAsO$_{1-x}$F$_x$ have found large values for TEP under ambient conditions [12]–[15]. The shapes of $S(T)$ curves are similar to those of strong correlated systems like layered cobalt oxides [16]–[18], while the resistivity is quite low. As in the cobaltite oxides, the iron oxypnictides involve multiple orbitals at the Fermi level and complex magnetic orderings correlated with the enhanced TEP, which makes them a promising candidate for TEP materials.

Here, we report our TEP investigation of SmFeAsO$_{0.85}$ at pressures up to 1.1 GPa. The magnitude of TEP decreases with increase in pressure. $T_c$ decreases monotonically with increase in pressure in contrast with the results for LaFeAsO$_{1-x}$F$_x$ [11]. $S(T)$ exhibits a broad peak, which decreases in magnitude and shifts to lower temperature under pressure. The temperature- and pressure-dependent behavior shows anomalous features that cannot be explained within the view of a simple metallic model. We attribute this unusual behavior to a resonant phonon-drag effect dominant in the normal state, in which the nesting of Fermi surfaces plays an important role.

2. Experimental

The polycrystalline SmFeAsO$_{0.85}$ samples, in which doping is achieved with oxygen vacancies instead of fluorine doping, were synthesized by the solid-state reaction method under high pressure [4, 5]. A thin-film heater is thermally attached to one end of the sample for generating the temperature gradient, which is measured with a chromel-constant differential thermocouple. The heater was turned on and off for a few cycles for signal averaging. The temperature difference is maintained below 0.1% of the base temperature over the entire temperature range.

New Journal of Physics 11 (2009) 025006 (http://www.njp.org/)
Figure 1. TEP $S$ (squares) and resistance (circles) versus temperature of SmFeAsO$_{0.85}$ at ambient pressure. The magnitude of $S$ grows into a broad peak at $T^*$, coexisting with a low resistance.

The data were corrected for the thermopower of the gold leads. The pressure was applied in a regular beryllium–copper clamped cell, with silicon oil inside a Teflon cup as the pressure-transmitting medium. A calibrated Manganin pressure gauge located inside the cell served to determine the pressure at room temperature.

3. Results and discussion

Figure 1 shows the temperature dependence of the thermopower and the resistance of SmFeAsO$_{0.85}$ at ambient pressures from 30–300 K. The resistance displays linear temperature dependence from $T_c \sim 53$ K onwards, and deviates from linear behavior above $\sim 180$ K. The shape of the $S$–$T$ curve displays an increase in magnitude with decreasing temperature and a broad peak of magnitude $S_p \sim 52 \mu$V K$^{-1}$ at $T^* \sim 110$ K, marked in figure 1, and then a rapid drop to zero at the onset of superconductivity. The normal state thermopower value is negative over the entire range of temperature, indicating electron-like charge carriers. These features are in good qualitative agreement with previous reports on LaFeAsO$_{1-x}$F$_x$ [12, 13]. It is also noteworthy that the temperature dependence of the TEP is unusual. The high-temperature upturn in the TEP does not follow a $(1/T)$ dependence, which is characteristic of the system opening of a gap at the Fermi level.

Figure 2 shows the evolution of the temperature dependence of the TEP from ambient pressure up to 1.1 GPa. The inset shows the low-temperature expansion around $T_c$. The superconducting $T_c$ is defined from the intersection of the two extrapolated lines, as shown in the inset of figure 2. The onset $T_c$, plotted in figure 3, decreases linearly with pressure, at a rate of $\sim -1.5$ K GPa$^{-1}$, which is consistent with the resistance measurements in the same compounds [21]. A previous report has revealed that pressure can either suppress or enhance $T_c$ depending on the doping level [22]. The width of the transition does not dramatically change with pressure, indicating a single-phase compound.

We now examine various models that might account for the observed TEP behavior. In general, TEP in metals is the sum of three different contributions: the diffusion term
Figure 2. Temperature dependence of the TEP at different pressures. The arrow indicates the direction of increasing pressure. The superconducting transition region is enlarged in the inset, and $T_c$ shift to lower temperature with increasing pressure.

Figure 3. Both superconducting transition temperature $T_c$ (squares) and temperature $T^\ast$ (circles) have linear pressure dependence, suggesting that the emergence of the superconducting phase should be related to the nesting of Fermi surfaces in the normal state. The dashed lines are guides to the eyes.

$S_{\text{diff}}$, the spin-dependent scattering term and the phonon-drag term $S_{\text{drag}}$. $S_{\text{diff}}$ is given by the Sommerfeld expression $S_{\text{diff}} \sim (k_B/e)(T/T_F)$, where $T_F$ is the Fermi temperature. Based on the Hall measurements, we can estimate that $T_F \sim 5000$ K [23]. Therefore, the TEP is strongly reduced by the factor $T/T_F$, which cannot explain the large measured value. This implies that the metallic diffusive contribution can be ignored in the measured thermopower.

One would tentatively ascribe the broad TEP peak to the Kondo mechanism, since our samples were synthesized with iron powders. In a classical magnetic impurity system, $S(T)$
exhibits a broad peak at $T_{\text{max}} \sim T_K$ (Kondo temperature), which is attributed to a ‘Kondo resonance’ in the electronic density of states near the Fermi level. However, $R(T)$ shows metallic behavior, and does not display any correlation with the broad peak in the $S(T)$ curve. On the other hand, $S(T)$ does not follow a logarithmic $T$-dependence, which is characteristic of the Kondo effect. We also find that the data cannot be fitted in a relation of power law dependence over any significant temperature range, which was suggested by the magnon-drag effect from the magnetic impurity \[24\]. When pressure is applied, we observe a trend toward a decrease of the magnitude of the peak, followed by a gradual shift of the characteristic temperature $T^*$ to a lower temperature, as shown in figure 3. However, within our pressure range, pressure cannot influence the sample impurity or the exchange coupling between the conduction electrons and the magnetic impurity. This observation excludes the possibility that the observed TEP behavior could be related to the magnetic impurities.

The phonon-drag effect is another candidate, which usually gives a peak structure in the temperature dependence of TEP between $\theta_d/10$ and $\theta_d/5$, where $\theta_d$ is the Debye temperature. For the FeAs superconductor, $\theta_d$ is estimated to be $\sim 170$ K from specific heat measurements \[25\]. Therefore, the conventional phonon-drag effect cannot explain the observed peak structure in $S(T)$ curves.

In the following, we will interpret the unusual temperature and pressure dependence of the TEP as due to resonant phonon drag in the presence of nesting of Fermi surfaces. The crystal structure of Fe-based compounds belongs to the tetragonal space group $P4/nmm$, and has multiple Fermi surface sheets around points $\Gamma(0, 0)$ and $M(\pi, \pi)$ (see the inset of figure 4) \[19, 20\]. Theoretical calculations and recent angle-resolved photoemission measurements have evidenced the significant nesting effect between points M and $\Gamma$, which leads to a spin density-wave instability \[6, 10, 19, 20\]. After shifting the hole Fermi surface around $\Gamma$ by a vector $Q = (\pi, \pi, 0)$, it will largely overlap with the electron Fermi surface around M. The resonant phonon drag involves only acoustic phonons with wave vector $Q$ in the resonant process from the nested Fermi surfaces. According to the calculations, a weak electron–phonon coupling strength cannot explain superconductivity in the iron-based superconductors. However, an average of electron–phonon coupling over the whole structure might be misleading. Recent density functional calculation indicated that the Fermi surface nesting effects in these compounds lead to a strong electron–phonon coupling \[26\].

In a simple model, the momentum selection rule is needed to absorb or emit the phonon with the corresponding energy $\hbar \omega_Q$. This results in a modified exponential relation

$$S \sim \exp(-E / k_B T)/T^2,$$

where $E \sim \hbar \omega_Q$ is the resonant phonon energy \[27\]–\[30\]. The temperature dependence of $S$ should show an upturn at low temperature and then a decrease at higher temperature, producing a peak in $S(T)$ at a characteristic temperature $T^* \sim \hbar \omega_Q/k_B$. Therefore, the TEP data are expected to follow a scaling behavior as a function of the reduced temperature $T/T^*$. In figure 4, the normalized TEP $S_{\text{nor}} = S/S_p$ under various pressures are plotted against the scaling parameter $T/T^*$. The three traces in figure 2 collapse nicely onto a single universal curve, in which logarithmic $S_{\text{nor}}T^2$ is approximately linear with $1/(T/T^*)$, consistent with resonant phonon-drag behavior. From the slope of the fit, we deduce the activation energy $E \sim 15.8$ meV, which is comparable with the theoretical estimate \[20\].

The application of pressure would suppress the nesting of the Fermi surface, playing a similar role as electron doping. Neutron diffraction shows that both the magnetic order and

New Journal of Physics 11 (2009) 025006 (http://www.njp.org/)
Figure 4. The plot of $-S_{\text{tot}} T^2$ versus $1/(T/T^*)$ under different pressures falls into a single scaling curve, predicted by equation (1). The linear fit to the data is indicated by a dashed line. The inset shows a schematic Brillouin zone and the calculated Fermi surface [6].

The structural distortion are suppressed with doping [9]. Theoretical calculations reveal that the electron-type Fermi surfaces around point M are ellipses. The shape of the two ellipses is modified by applying pressure [31]. Under pressure, the mismatch between electron and hole Fermi surfaces is enhanced, and the nesting effect is suppressed. In such a case, the application of pressure would decrease the magnitude of TEP, and shift $T^*$ to lower temperatures, as shown in figure 3. The scaling relation shown in figure 4 emphasizes that increasing pressure drives both softening of the resonant phonon mode and suppression of the TEP amplitude.

Finally, we would like to point out another possible combined effect of the spin and orbital degeneracy on the TEP in iron oxypnictides. In materials with strong interactions, the spin and orbital degrees of freedom are predicted to produce a large contribution of the Heikes form [17, 32],

$$S = \frac{\mu}{eT} = -\frac{\sigma}{e},$$

where $\mu$ is the chemical potential, and $\sigma = k_B \ln(g_s g_o)$ with $g_s$ and $g_o$ being the spin and orbital degeneracies, respectively. Theoretical calculations have shown that TEP is strongly enhanced by spin and orbital degeneracy and reaches a maximum at a characteristic temperature [17, 33]. For the FeAs superconductors, theoretical calculations have revealed that the dominant states at the Fermi level come from all five Fe 3d orbitals, and the compound in the normal state is a strongly correlated metal [34]. Application of pressure would increase the band width with decreasing the strong effect of Coulomb interaction and thus suppress the enhanced TEP.

4. Conclusion

In summary, the temperature dependence of TEP under pressure was measured on SmFeAsO$_{0.85}$ superconductors. The magnitude of TEP develops a broad peak coupled with a low electrical
resistivity. The temperature and pressure dependence of TEP in the normal state can be explained by the resonant phonon scattering between the hole and electron pockets in the presence of nesting of the Fermi surfaces. To further understand the enhanced $S$, a theory including orbital and spin degrees of freedom is possibly needed.

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