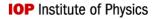
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# Nernst effect of a new iron-based superconductor $LaO_{1-x}F_xFeAs$

Z W Zhu $^1$ , Z A Xu $^{1,4}$ , X Lin $^1$ , G H Cao $^1$ , C M Feng $^2$ , G F Chen $^3$ , Z Li $^3$ , J L Luo $^3$  and N L Wang $^3$ 

<sup>1</sup> Department of Physics, Zhejiang Unviersity, Hangzhou 310 027, People's Republic of China

<sup>2</sup> Test and Analysis Center, Zhejiang University, Hangzhou 310 027, People's Republic of China

<sup>3</sup> Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100 080, People's Republic of China

E-mail: zhuan@zju.edu.cn

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**Abstract.** We report the first Nernst effect measurement on a new iron-based superconductor  $LaO_{1-x}F_xFeAs$  (x=0.1). In the normal state, the Nernst signal is negative and very small. Below  $T_c$ , a large positive peak caused by vortex motion is observed. The flux flowing regime is quite large compared to the conventional type-II superconductors. The sharp decrease in the Nernst signal at the depairing magnetic field  $H_{c2}$  is not obvious even for temperatures close to  $T_c$ . Furthermore, a clear deviation of the Nernst signal from normal state background and an anomalous suppression of off-diagonal thermoelectric current in the normal state between  $T_c$  and 50 K are observed. We argue that this anomaly in the normal state Nernst effect could correlate with the spin-density wave (SDW) fluctuations although the contribution of superconducting fluctuations cannot be excluded.

<sup>&</sup>lt;sup>4</sup> Author to whom any correspondence should be addressed.

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#### 1. Introduction

Since the discovery of superconductivity in the quaternary oxypnictides LaOFeP [1] and LaONiP [2], a new family of high- $T_c$  superconductors featuring iron-based layered structures have emerged [3]–[10]. A non-conventional superconducting mechanism has been proposed based on tunneling spectroscopy [11], thermodynamics measurements [12] and first-principles calculations [13, 14]. A recent band structure calculation indicates that the Fermi surface (FS) of  $\text{LaO}_{1-x}F_x\text{FeAs}$  has five sheets [13]–[15]: two cylinders associated with electron-type around the zone edge M-A line of the Brillouin zone, two hole-like cylinders along the  $\Gamma-Z$  line, and a hole-like closed pocket centered at the Z point. The calculations also indicate that LaOFeAs is basically characterized as low density electron carriers doped on top of a band insulator with filled  $d^6$  valence orbitals [13]. F doping can reduce the hole-like sheet and enhance the electron-like FS. Hall effect measurements [4, 5] have confirmed the electron-type conductivity with a charge carrier density as low as  $1.8 \times 10^{-21}$  cm<sup>-3</sup> for F-doped systems, even lower than that of high- $T_c$  cuprates.

The low dimensionality and low charge carrier density in this system remind us of the analogy to high- $T_{\rm c}$  cuprates. Hence, the superfluid density  $n_{\rm s}$  in this system should also be very small compared to that of typical metal or alloy superconductors. It has been proposed that the small superfluid density results in a small phase stiffness energy and the Meissner state is readily destroyed by the strong phase fluctuations [16]. In the case of high- $T_{\rm c}$  cuprates, the correlation between the  $T_{\rm c}$  and superfluid density demonstrated by the so-called 'Uemura plot' [17] strongly implies such a phase-disorder scenario in which the superconducting transition at  $T_{\rm c}$  is driven by the loss of long-range phase coherence rather than 'gap closing'. Furthermore, persistence of the enhanced Nernst signals due to the vortex excitations at temperatures rather high above  $T_{\rm c}$  provides strong evidence for the spontaneous appearance of vortex excitations above  $T_{\rm c}$  [18, 19].

The Nernst effect is a magneto-thermoelectric effect defined as the appearance of a transverse electric field  $E_y$  in response to a temperature gradient  $\nabla T \parallel x$ , in the presence of a perpendicular magnetic field  $H \parallel z$  and under open circuit conditions. The Nernst effect is usually small in usual metals because of the so-called 'Sondheimer cancellation' [20]. It is known that the magnetic ordering and spin fluctuations can cause enhanced Nernst signals [21, 22]. For a type-II superconductor (in the vortex-liquid state), vortices can be driven down by the temperature gradient with velocity v along the x-axis direction. Moving vortices can produce a transverse electric field according to the Josephson equation:  $E_y = B \times v$ , which is also called the vortex Nernst effect. Thus, the vortex Nernst effect is a highly sensitive probe for detecting vortex motion.

Here, we report the first Nernst effect measurement on a newly discovered iron-based superconductor  $\text{LaO}_{1-x}F_x\text{FeAs}$  (x=0.1). A broad regime below  $T_c$  featuring a large positive vortex Nernst signal is observed, which indicates that the flux-flowing regime is quite large compared to that of the conventional type-II superconductors. However, a clear anomalous depression of off-diagonal thermoelectric current below 50 K in the normal state is observed and its origin is discussed.

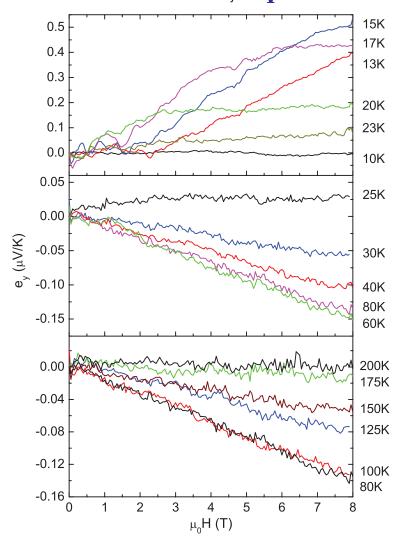
#### 2. Experimental

The polycrystalline samples were prepared by solid-state reaction using LaAs, Fe<sub>2</sub>O<sub>3</sub>, Fe and LaF<sub>3</sub> as the starting materials. The sample preparation details can be found in [4]. The powder x-ray diffraction patterns indicate that the resultant is single phase and all the diffraction peaks can be well indexed based on the tetragonal ZrCuSiAs-type structure with the space group P4/nmm. The structure characterization was reported in [4]. The nominal composition is LaO<sub>0.9</sub>F<sub>0.1</sub>FeAs according to the starting materials. The resistivity measurement shows a sharp superconducting transition and  $T_c$  (the onset temperature in the resistive transition) is 26 K. The residual resistance ratio (RRR) which is defined as R(300 K)/R(28 K) is 11.6, indicating a good sample quality.

The Hall effect was measured by scanning the magnetic field at fixed temperatures. The thermoelectric properties were measured by a steady-state technique. The temperature gradient used for the thermoelectric measurements, determined by a pair of differential type E thermocouples, was around  $0.5 \, \mathrm{K} \, \mathrm{mm}^{-1}$ . All the measurements were performed in a Quantum Design PPMS-9 system. The Nernst signal  $e_y$  is defined as  $e_y \equiv \frac{E_y}{|\nabla T|}$ . The Nernst signal was measured at positive and negative field polarities, and the difference of the two polarities was deduced in order to remove any thermopower contribution.

#### 3. Results and discussion

Traces of the Nernst signal as a function of magnetic field up to  $\mu_0 H$  of 8 T are displayed in figure 1 at various temperatures. Above  $T_c$  of about 26 K, the Nernst signal  $(e_v)$  is negative, small and linear as a function of magnetic field. Below  $T_c$ , the magnitude of the Nernst signal increases sharply, and becomes nonlinear with magnetic field, which is a typical feature of the vortex Nernst signal. At 10 K, vortices form a solid lattice and are pinned so that the vortex Nernst signal is nearly zero. At T = 13 and 15 K, the vortex lattice melts at a certain magnetic field  $H_{\rm m}$  and the vortex Nernst signals appear for  $H > H_{\rm m}$ . For T > 17 K, such a melting transition of vortex lattice is no longer observed just because the solid vortex lattice is not formed due to thermal fluctuations. Probably because of the polycrystalline sample, the vortex motion is not uniform and enhanced noise is observed which might be caused by vortex jumping. It is interesting that the traces of the vortex Nernst signals below  $T_c$  are similar to those of high- $T_c$ cuprates, i.e. the vortex liquid state regime is quite large and the solid vortex lattice is hard to form. Another surprising feature is that the depairing magnetic field  $H_{c2}$  cannot be defined in the  $e_v$  versus H curves even for temperatures near  $T_c$ . The magnetoresistance measurement performed on the same batch of  $LaO_{1-x}F_xFeAs$  (x = 0.1) samples was reported in [4] and  $H_{c2}(T)$  was determined from the onset transition of resistance for temperatures close to  $T_c$ .  $\mu_0 H_{c2}(0)$  was estimated to be about 55 T and  $H_{c2}(T)$  drops down to zero quickly when T approaches  $T_{c0}$ . However, the Nernst effect study indicates that  $H_{c2}$  might not drop to zero at  $T_c$ 

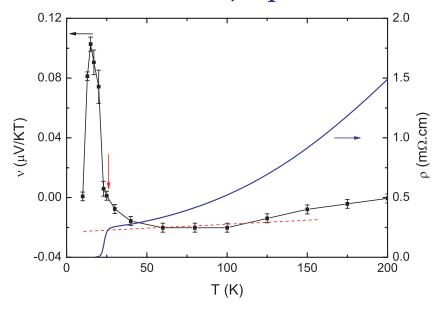


**Figure 1.** Magnetic field dependence of the Nernst signal  $(e_y)$  at different temperatures.

and the superconducting transition could be driven by the loss of long-range phase coherence of Cooper pairs, just as in the high- $T_c$  superconducting cuprates [23]. Such a behavior is in contrast to conventional layered superconductors such as NbSe<sub>2</sub> [24, 25].

The temperature dependence of the Nernst coefficient,  $\nu(T)$ , together with its resistivity,  $\rho(T)$ , is shown in figure 2. The Nernst coefficient is defined as the initial slope of  $e_y$ –H curves for  $T < T_c$ . The normal state Nernst coefficient is very small and shows weak temperature dependence. However,  $\nu$  starts increasing below 50 K, changes sign just above  $T_c$  and then shows a step-like sharp increase at  $T_c$ . Below  $T_c$ , it reaches a peak around 15 K. The peak value is around  $0.10 \,\mu\text{V} \,(\text{K T})^{-1}$ , smaller than the vortex Nernst signal observed in high- $T_c$  superconductors, probably because of the polycrystalline sample in this study. A sharp increase in  $\nu(T)$  occurs at  $T_c$ , in contrast to the smooth change of  $\nu(T)$  at  $T_c$  in high- $T_c$  superconductors. However, for T as high as 50 K above  $T_c$ ,  $\nu(T)$  already starts deviating from the negative background and increases gently. In high- $T_c$  cuprates, a similar enhancement of the Nernst signal has already been observed and it has been interpreted as evidence for vortex excitations





**Figure 2.** Temperature dependence of the Nernst coefficient. The resistivity measured under zero magnetic field is also shown. The arrow indicates  $T_c$  of 26 K. The dashed line is a guide to the eye.

above  $T_c$ . A phase-disorder scenario based on the Nernst effect measurements is proposed for high- $T_c$  cuprates [18, 19]. Because these iron-based new superconductors share a similarity in low dimensionality and low superfluid density with high- $T_c$  cuprates, it is plausible to ascribe the deviation of the Nernst signal from the normal state background to the vortex excitations existing in a region up to 20 K above  $T_c$ . However, as is discussed hereafter, we argue that this anomaly change in the Nernst signal between  $T_c$  and 50 K might correlate with the spin-density wave (SDW) fluctuations rather than superconducting phase fluctuations.

In order to explore the nature underlying this anomaly in the Nernst effect, it is useful to isolate the off-diagonal thermoelectric (Peltier conductivity) term from the Nernst signal by measuring the Hall effect and Seebeck effect separately. The temperature dependence of the Hall coefficient ( $R_{\rm H}$ ) and thermopower (S) were measured for the same sample under the magnetic field ( $\mu_0 H$ ) of 8 T, and they are shown in figures 3(a) and (b). Their values in the normal state are consistent with previous reports [4, 5, 26] and the negative signs mean that the dominant charge carrier is electron-like. At  $T_{\rm c}$ , both  $R_{\rm H}$  and S drop to zero sharply. It is very interesting that the profile of S versus T curves is similar to that of the low charge density metals like underdoped high- $T_{\rm c}$  cuprates and Na $_{\rm x}$ CoO2, except for the negative sign.

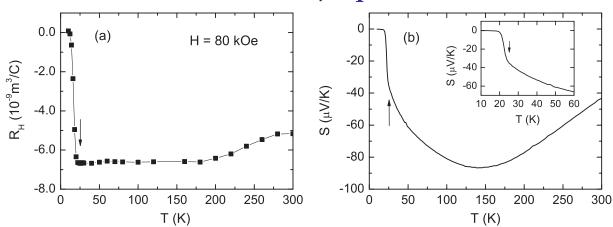
The normal state Nernst signal consists of two terms, viz,

$$e_{v} = \rho \alpha_{xv} - S \tan \theta, \tag{1}$$

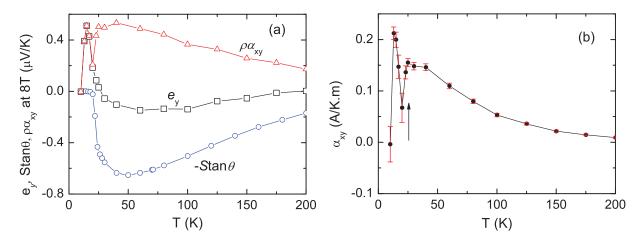
where  $\alpha_{xy}$  is the off-diagonal Peltier coefficient,  $S = \frac{\alpha}{\sigma}$  the thermopower ( $\sigma$  the diagonal conductivity and  $\alpha$  the diagonal Peltier coefficient),  $\rho$  the resistivity and  $\tan \theta$  the Hall angle. For usual simple metals, the two terms in equation (1) cancel each other, which is the so-called 'Sondheimer cancellation' [20]. The second term  $S \tan \theta$  is calculated based on the S and Hall effect data, which is shown in figure 4(a). By comparing  $e_y$  with  $S \tan \theta$  in figure 4(a), it becomes clear that the Nernst signal  $e_y$  is about one fifth of  $S \tan \theta$ , which means the 'Sondheimer







**Figure 3.** (a) Temperature dependence of the Hall coefficient  $(R_{\rm H})$  measured under a magnetic field  $(\mu_0 H)$  of 8 T. (b) Temperature dependence of thermopower (S). The inset shows an enlarged plot of S versus T around  $T_{\rm c}$ .



**Figure 4.** (a) Temperature dependence of the Nernst signal  $(e_y)$ , the product term  $S \tan \theta$  and off-diagonal Peltier term  $\rho \alpha_{xy}$ . (b) Temperature dependence of the off-diagonal Peltier coefficient  $\alpha_{xy}$ . The arrow indicates  $T_c$  of 26 K.

cancellation' is not complete<sup>5</sup>. However, the multiband character does not significantly enhance the Nernst signals in the normal state. In other words, only one type (electron-type) of charge carrier is dominant in the Nernst effect of the F-doped  $\text{LaO}_{1-x}F_x\text{FeAs}$ . The first term  $\rho\alpha_{xy}$  in equation (1) is calculated by subtracting  $e_y$  from the product— $S \tan \theta$ , which is shown in figure 4(b). It can be found that the off-diagonal thermoelectric current  $\alpha_{xy}$  increases with decreasing temperature at high temperatures, but it saturates around 50 K and starts decreasing quickly as the temperature nears  $T_c$ . Around and below  $T_c$ , there are three terms which comprise  $e_y$ , viz,

$$e_{v} = \rho \alpha_{xv}^{s} + \rho \alpha_{xv}^{n} - S \tan \theta, \tag{2}$$

<sup>&</sup>lt;sup>5</sup> In the case of hole-doped high- $T_c$  cuprates,  $e_y$  is only about 1/10 of  $S \tan \theta$ , i.e. the Sondheimer cancellation is nearly complete.

where  $\rho \alpha_{xy}^s$  denotes the contribution of vortices to the Nernst signals, and  $\rho \alpha_{xy}^n$  denotes the contribution from the normal state excitations. Below  $T_c$ ,  $\rho \alpha_{xy}^n$  should drop to zero quickly, and  $\rho\alpha_{xy}^s$  shows a large positive peak. However, the  $\rho\alpha_{xy}^n$  term starts decreasing at temperatures far above  $T_c$ , which means a change in the electron state below 50 K. The off-diagonal Peltier coefficient  $(\alpha_{xy})$  shows a similar profile to that of  $\rho\alpha_{xy}^n$ . It has been proposed that the undoped parent compound LaOFeAs undergoes an SDW transition around 134 K and the F doping will suppress the SDW transition, and finally give way to superconductivity [27]-[29]. In F-doped superconductors, there is evidence to support the existence of magnetic fluctuations. The variation of the temperature dependence of the resistivity with F content shows that the SDW transition is gradually suppressed by F doping, and a competition of the SDW order and the superconductivity order was proposed [27]. Recently, Hosono and co-workers reported that a pseudogap of 0.1 eV in  $LaO_{1-x}F_xFeAs$  was detected by photoemission and they attributed the origin of the pseudogap to strong spin fluctuations [30]. They also found that the spin fluctuation shows a maximum at 5% F doping by dc magnetization measurements [31]. The slow increase in  $e_v$  above 50 K might be caused by the residual magnetic fluctuations, and therefore we suggest that the sharp decrease in  $\rho \alpha_{xy}^n$  below 50 K could result from the suppression of magnetic fluctuations. In other words, there is a kind of 'precursor state' between T<sub>c</sub> and 50 K in which the magnetic fluctuations are strongly suppressed. Such a suppression of magnetic fluctuations should be detected by other probes like nuclear magnetic resonance (NMR) measurement. Recent NMR measurements by Nakai et al [32] indicated that the  $1/TT_1$ of <sup>75</sup>As in a LaO<sub>0.96</sub>F<sub>0.4</sub>FeAs sample shows a Curie-Weiss behavior down to 30 K, suggesting the development of SDW spin fluctuations with decreasing temperature. Then it is suppressed below 30 K. They concluded that superconductivity emerges when magnetic ordering is suppressed. We suggest that the sharp suppression of the off-diagonal Peltier current  $\alpha_{xy}$  and of  $1/TT_1$  just above  $T_{\rm c}$  should have the same origin, i.e. both should result from the suppression of SDW fluctuations. It is an interesting open question whether this 'precursor state' is indeed crucial for the emergence of 'high- $T_c$ ' superconductivity in this system.

#### 4. Conclusion

We report the first Nernst effect measurement on a new iron-based superconductor  $LaO_{1-x}F_xFeAs$  (x=0.1). In the normal state, the Nernst signal is negative and very small, which means that the 'Sondheimer cancellation' is incomplete and the normal state transport is mainly dominated by the electron-like band. Below  $T_c$  a large positive peak related to vortex motion is observed. The flux flowing regime is very large, and there is no obvious change in the Nernst signals at  $H_{c2}$  which is defined from the magnetoresistance, in contrast to the conventional type-II superconductors with layered structure. Furthermore, a clear deviation of the Nernst signal from normal state background and an anomalous suppression of off-diagonal thermoelectric current between  $T_c$  and 50 K are observed. We argue that this anomaly in the normal state Nernst effect could correlate with the SDW fluctuations.

#### **Acknowledgments**

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