Physica C 469 (2009) 566–574

Contents lists available at ScienceDirect

Physica C

journal homepage: www.elsevier.com/locate/physc

High-field phase-diagram of Fe arsenide superconductors

Y.J. Jo [a], J. Jaroszynski [a], A. Yamamoto [a], A. Gurevich [a], S.C. Riggs [a], G.S. Boebinger [a], D. Larbalestier [a], H.H. Wen [b], N.D. Zhigadlo [c], S. Katrych [c], Z. Bukowski [c], J. Karpinski [c], R.H. Liu [d], H. Chen [d], X.H. Chen [d], L. Balicas [a,*]

[a] National High Magnetic Field Laboratory, Florida State University, Tallahassee-FL 32310, USA
[b] Institute of Physics, Chinese Academy of Sciences, Beijing 100190, PR China
[c] Laboratory for Solid State Physics, ETH Zürich, CH-8093 Zürich, Switzerland
[d] Hefei National Laboratory for Physical Science a Microscale and Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, PR China

ARTICLE INFO

Article history:
Available online 19 March 2009

PACS:
74.25.–q
74.25.Ha
74.25.Op
74.70.Dd

Keywords:
High magnetic fields
Electrical transport
Torque magnetometry
Superconducting phase-diagram

ABSTRACT

Here, we report an overview of the phase-diagram of single-layered and double-layered Fe arsenide superconductors at high magnetic fields. Our systematic magneto-transport measurements of polycrystalline $SmFeAsO_{1-x}F_x$ at different doping levels confirm the upward curvature of the upper critical magnetic field $H_{c2}(T)$ as a function of temperature T defining the phase boundary between the superconducting and metallic states for crystallites with the ab planes oriented nearly perpendicular to the magnetic field. We further show from measurements on single-crystals that this feature, which was interpreted in terms of the existence of two superconducting gaps, is ubiquitous among both series of single- and double-layered compounds. In all compounds explored by us the zero temperature upper critical field $H_{c2}(0)$, estimated either through the Ginzburg–Landau or the Werthamer–Helfand–Hohenberg single gap theories, strongly surpasses the weak-coupling Pauli paramagnetic limiting field. This clearly indicates the strong-coupling nature of the superconducting state and the importance of magnetic correlations for these materials. Our measurements indicate that the superconducting anisotropy, as estimated through the ratio of the effective masses $\gamma = (m_c/m_{ab})^{1/2}$ for carriers moving along the *c*-axis and the *ab-planes*, respectively, is relatively modest as compared to the high-T_c cuprates, but it is temperature, field and even doping dependent. Finally, our preliminary estimations of the irreversibility field $H_m(T)$, separating the vortex-solid from the vortex-liquid phase in the single-layered compounds, indicates that it is well described by the melting of a vortex lattice in a moderately anisotropic uniaxial superconductor.

1. Introduction

The iron-based oxypnictides represent a novel class of superconductors which, with the exception of the cuprates, have the highest known superconducting transition temperature T_c [1,2]. Several series of these compounds have been synthesized in the last year, but throughout this manuscript we will mostly focus on the properties of the single-layered oxypnictide $LnFeAsO_{1-x}F_x$ (*Ln* is a lanthanide), or the so-called 1111 compounds, and on the $AEFe_2As_2$ (*AE* is an alkali-earth metal), or the so-called 122 compounds.

Both electric transport measurements and electronic band structure calculations suggest that undoped oxypnictides are semimetals [3]. There is an approximate nesting between the hole Fermi surface (FS) centered at the Γ point and the electron FS centered at the M point, which may lead to a spin-density wave (SDW) like instability state observed at low temperatures [4,5]. According to neutron diffraction studies, the magnetic structure of undoped pnictides is composed of antiferromagnetically coupled ferromagnetic chains [6]. Superconductivity in the 1111 or electron doped compounds would occur when part of the Fe^{2+} ions are replaced by Fe^{+}, which is expected to suppress the antiferromagnetic instability.

Several superconducting pairing mechanisms based on the multi-band nature of these compounds have been proposed. Dai et al. [7] suggested a spin-triplet pairing mechanism with even parity due to ferromagnetic spin fluctuations between electrons in different orbitals. Lee and Wen [8] argued that the strong Hunds rule ferromagnetic interaction in Fe pnictides can lead to a pairing instability in the spin-triplet *p*-wave channel in the weak-coupling limit, so that the superconducting gap would have nodal points on the two-dimensional Fermi surfaces. While Lee et al. suggested that because of the frustrating pairing interactions among the electron and the

* Corresponding author.
E-mail address: balicas@magnet.fsu.edu (L. Balicas).

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doi:10.1016/j.physc.2009.03.010

hole fermi-surface pockets, a $s + id$ pairing state with broken time reversal symmetry could be favored [9]. Perhaps, the model that is currently more widely accepted is the so-called extended $s^{\pm}$-wave model, which predicts a π shift between the order parameters on the hole and the electron Fermi-surface sheets [4]. In this scenario, the unconventional pairing mechanism is mediated by antiferromagnetic spin fluctuations. In fact, inelastic neutron scattering in the $Ba_{0.6}K_{0.4}Fe_2As_2$ compound reveals the emergence of a localized resonant magnetic excitation below the superconducting transition temperature [10]. This type of excitation is expected for a superconducting order parameter which has opposite signs in different parts of the Fermi surface as in the $s^{\pm}$ scenario. A general overview of the different pairing scenarios in oxypnictides is given by Mazin and Schmalian in this volume [11].

It is interesting to mention a recent photoemission report [12] claiming the existence of an underlying electronic (π,π) order in $Ba_{1-x}K_xFe_2As_2$, previously seen in the cuprates [13] and claimed to be perhaps at the origin of the observed small Fermi-surface pockets, as also seen in underdoped $YBa_2Cu_3O_{7+\delta}$ [14]. Thus the coexistence and non-trivial interplay of different order parameters seems to be an essential ingredient for high temperature superconductivity, in either the cuprates or the oxypnictides.

As for the existence of multiple superconducting gaps owing to the multi-band nature of the Fe arsenides the situation is still somewhat inconclusive. Several point contact spectroscopy studies yield contradictory results, with evidence for both single [15] and multi-gap superconductivity [16]. The latter does not invalidate the multi-band pairing scenario but rather indicates that the gaps of approximately equal magnitudes may reside on different disconnected sheets of the Fermi surface. At the same time, detailed local magnetization measurements via Hall probe magnetometry [17], photoemission [18,19] and neutron [10] measurements are consistent with a two-gap superconducting scenario for the 122 compounds.

The exact shape of the $H - T$ superconducting phase-diagram can help distinguish between different theoretical scenarios. Moreover, magneto-transport measurements under strong magnetic field can also determine the temperature dependence of the irreversibility field, which is one of the key parameters quantifying the potential of the oxypnictides for future power applications. However, it is already clear [20,21] that these compounds are characterized by tremendously large upper critical fields, requiring very high magnetic field techniques to explore the overall complexity of their superconducting phase-diagram. This is in turn a very encouraging result for applied superconductivity.

There have been already several reports on the physical properties of the Fe arsenides at very high magnetic fields. Two relevant reports, provide (i) evidence for two-gap superconductivity to explain the upward curvature of the upper critical field as a function of temperature for fields along the c-axis direction in 1111 compounds [20,21], and (ii) an evidence that the $H_{c2}(T)$ of the 122 compounds is weakly anisotropic despite the two-dimensional character of their Fermi surface, thus indicating that a strong anisotropy may not be instrumental for the high-T_c superconductivity in the Fe arsenides [22].

One of the greatest challenges at the moment is to achieve the synthesis of high quality single-crystals, and to explore their phase diagrams at very high fields not only by traditional magneto-tranport measurements but also by thermodynamic means. Here, we provide a brief overview of our initial efforts in this direction.

2. Experimental

Polycrystalline samples with nominal composition $SmFeAsO_{1-x}F_x$ were synthesized in Heifei by conventional solid state reaction using high-purity SmAs, SmF_3, Fe and Fe_2O_3 as starting materials. SmAs was obtained by reacting Sm chips and As pieces at 600 °C for 3 h and then 900 °C for 5 h. The raw materials were thoroughly ground and pressed into pellets. The pellets were wrapped in Ta foil, sealed in an evacuated quartz tube, and finally annealed at either 1160 °C or 1200 °C for 40 h. X-ray diffraction (XRD) pattern for a sample annealed at 1160 °C did reveal trace amounts of the impurity phase SmOF [2].

For the growth of $SmFeAsO_{1-x}F_x$ single-crystals at ETH [23], FeAs, Fe_2O_3, Fe and SmF_3 powders were used as starting materials and NaCl/KCl was used as flux. The precursor to flux ratio varied between 1:1 and 1:3. Pellets containing precursor and flux were placed in a BN crucible inside a pyrophyllite cube with a graphite heater. Six tungsten carbide anvils generated pressure on the whole assembly (3 GPa was applied at room temperature). While keeping pressure constant, the temperature was ramped up within 1 h to the maximum value of 1350–1450 °C, maintained for 4–10 h and decreased in 5–24 h to room temperature for the crystal growth. Then pressure was released, the sample removed and in the case of single-crystal growth NaCl/KCl flux was dissolved in water. Below, in our discussion concerning the irreversibility line in the 1111 compounds, we include some measurements previously reported by us in Ref. [21] in $NdFeAsO_{0.7}F_{0.3}$ single-crystals (see Ref. [24] for the details concerning the sample synthesis) which were originally used to extract the boundary between metallic and superconducting states at high fields.

The results reported here on 122 compounds were measured on single-crystals of $BaFe_{2-x}Co_xAs_2$ and $Ba_{1-x}K_xFe_2As_2$ grown at Hefei by the self-flux method. In order to avoid contamination from incorporation of other elements into the crystals, FeAs was chosen as self-flux. FeAs and CoAs powder was mixed together, then roughly grounded. The Ba pieces were added into the mixture. The total proportion of Ba:(2-xFeAs+xCoAs) is 1:4. For more details, see Ref. [25]. A similar procedure was used to synthesize single-crystals of $Ba_{1-x}K_xFe_2As_2$.

For electrical transport measurements, polycrystalline samples were cut into bar shaped pieces. Contacts for the standard four probe measurements were made by attaching gold wires with silver epoxy. Contacts in single-crystals were made by the focused-ion-beam technique as described in Ref. [24]. For the magnetic torque measurements a $SmFeAsO_{0.8}F_{0.2}$ single-crystal was attached to the tip of a piezo-resistive micro-cantilever which was itself placed in a rotator inserted into a vacuum can. Changes in the resistance of the micro-cantilever associated with its deflection and thus a finite magnetic torque τ was measured via a Wheatstone resistance bridge. The ensemble was placed into a ^{4}He cryostat coupled to a resistive 35 T dc magnet. For the transport measurements we used a combination of pulsed and continuous fields including the 45 T hybrid magnet of the National High Magnetic Field Lab in Tallahassee.

3. Results and discussion

3.1. Polycrystalline material

In Fig. 1 we show typical resistivity curves for polycrystalline $SmFeAsO_{1-x}F_x$ as a function of temperature under several values of magnetic field and for four nominal doping levels, $x = 0.1$, 0.15, 0.18, and 0.2, [26] respectively. The first striking feature is that one does not see a very pronounced broadening of the superconducting transition under strong fields, in marked contrast with the thermally-activated broadening seen in the cuprates [27]. Although, both polycrystalline and single crystalline $SmFeAsO_{1-x}F_x$ samples exhibit clear signatures for thermally-activated flux-flow [21] indicating the existence of a vortex-liquid state over a broad range of temperatures and magnetic fields. The broad peak seen in the resistivity above the superconducting state for $x = 0.1$