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Upper critical fields up to 60 T and the vortex matter phase diagram of arsenic-deficient $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$

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Abstract. We report resistivity, magnetization and upper critical field $B_{c2}(T)$ data for arsenic deficient $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$ in a wide temperature and high field range up to 60 T. These disordered samples exhibit a slightly enhanced transition temperature of T_c = 29.0 K and a significantly enlarged slope dB_{c2}/dT = –5.4 T/K near T_c. The high-field $B_{c2}(T)$ data obtained from resistance measurements in pulsed magnetic fields follow up to about 30 T the WHH (Werthamer-Helfand-Hohenberg) curve for the orbital limited upper critical field, but show a clear flattening above 30 T. This flattening evidences Pauli limiting behavior (PLB) with $B_{c2}(0)$ ≈ 63 T. We compare our results with $B_{c2}(T)$ data reported in the literature for clean and disordered samples. Whereas clean samples show no PLB for fields below 70 T as measured so far, the hitherto unexplained flattening of $B_{c2}(T)$ for applied fields H||ab observed for several disordered closely related systems is interpreted as a manifestation of PLB. The influence of the arsenic vacancies in $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$ on the vortex matter phase diagram is studied by magnetization measurements on bulk samples.

1. Introduction

The recently discovered FeAs based superconductors [1] exhibit high superconducting transition temperatures T_c up to 57 K and remarkably high upper critical fields $B_{c2}(0)$ exceeding often 70 T. Many basic properties of these novel superconductors and the underlying pairing mechanism are still not well understood. A study of the upper critical field and, in particular, investigations on disordered FeAs superconductors are of large interest since for an unconventional pairing both T_c and dB_{c2}/dT at T_c are expected to be suppressed by the introduction of controlled disorder. Here, the temperature dependence of the upper critical field, $B_{c2}(T)$, of arsenic-deficient (AD) $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$ samples is studied in fields up to 60 T. Furthermore, the vortex matter phase diagram of our AD sample is investigated by magnetization measurements and compared with that of a stoichiometric reference sample. Finally, the critical current density j_c within the grains of these samples is estimated from magnetization data and compared with j_c data obtained from transport measurements.

2. Experiments

Polycrystalline samples of $LaO_{0.9}F_{0.1}FeAs$ were prepared by the standard solid state reaction method

[2,3]. Some samples have been wrapped in a Ta foil during the final annealing procedure. Ta acts as an As getter at high temperatures forming a solid solution of about 9.5 at.% As in Ta. This leads to an As loss in the samples resulting in a As/Fe ratio of about 0.9 according to an energy-dispersive x-ray spectroscopy (EDX) analysis. A powder-x-ray diffraction study with a Rietveld refinement of the main phase yields enhanced lattice constants of a = 0.4028 nm and c = 0.8724 nm for the AD sample compared with a = 0.4020 nm and c = 0.8696 nm for the reference sample.

The electrical resistance was measured for plate-like samples using the standard four-point method. These measurements were done in a Physical Property Measurement System (PPMS, Quantum Design) in fields up to 14 Tesla. In addition, resistance measurements were performed in the pulsed-field facilities of the IFW Dresden and the FZ Rossendorf up to 50 T and 60 T, respectively. Gold contacts (100 nm thick) were made by sputtering in order to provide a low contact resistivity and to avoid possible heating effects in the pulsed field measurements. Magnetization measurements were carried out in magnetic fields up to 7 T using a SQUID magnetometer (MPMS, Quantum Design). The critical current of the investigated samples was determined both from magnetization and from transport measurements.

3. Results and discussion

The AD samples exhibit an about three times larger resistivity above T_c at 31 K than the clean reference samples which is due to enhanced disorder in the FeAs layer. Nevertheless, the AD samples have, with T_c = 28.5 K, a higher T_c than stoichiometric reference samples (T_c = 27.7 K) [3].

3.1. Upper critical field

The temperature dependence of the resistance of a typical AD sample is shown in figure 1 for applied fields up to 50 T. The upper critical field B_{c2} was determined from the onset of superconductivity at 90% of the resistance R_N in the normal state. The so defined B_{c2} corresponds to the highest upper critical field B_{c2}^{ab} which is related to those grains within the polycrystalline sample oriented with their *ab*-planes along the applied field.

In figure 2, the temperature dependence of B_{c2}^{ab} of our AD sample obtained from pulsed field measurements is shown together with B_{c2} data reported for a clean reference sample [4]. The large slope dB_{c2}/dT= -5.4 T/K at T_c of our AD sample points to strong impurity scattering in accord with its enhanced resistivity at 30 K. For the clean sample [4] shown in figure 2,

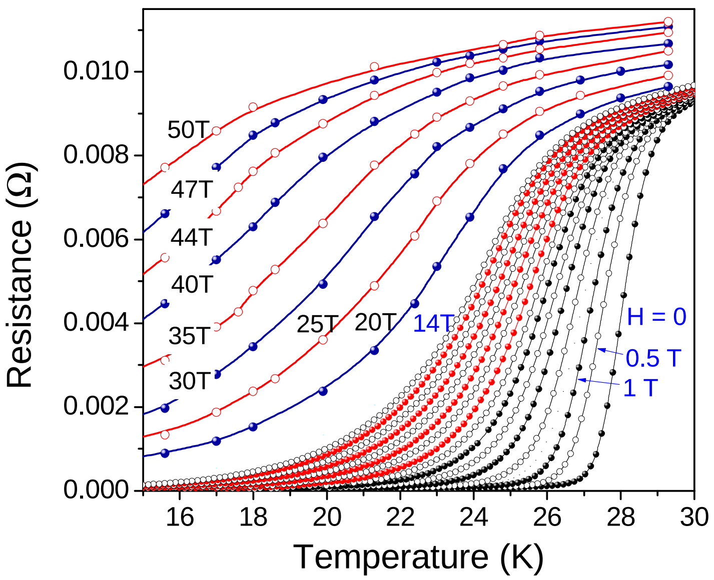

Figure 1. Temperature dependence of the resistance of a AD sample from both DC and pulsed field measurements.

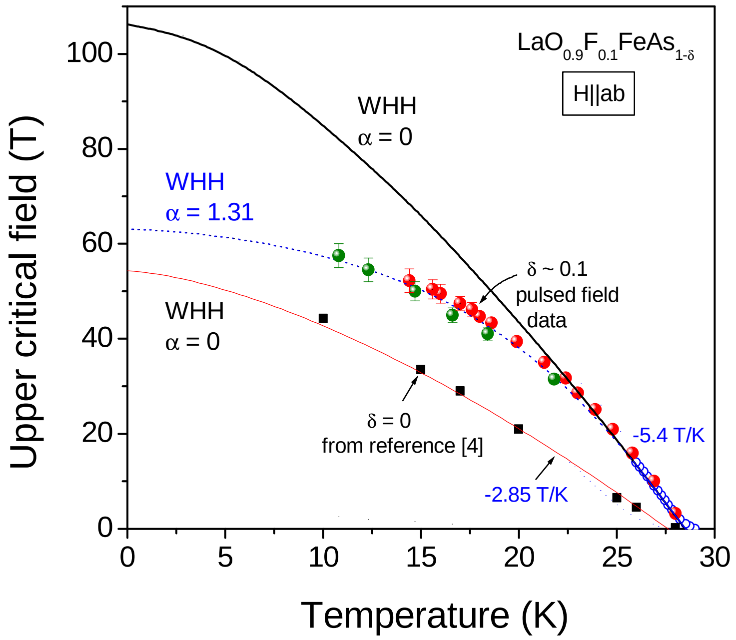

Figure 2. Temperature dependence of B_{c2}^{ab} for $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$. Data for the AD sample from DC (○) and pulsed field measurements (● - IFW Dresden, ● - FZD) in comparison with data (■) for a clean reference sample [4]. Solid lines: WHH model without PLB. Dotted line: $B_{c2}^{P}(T)$ for α = 1.31 (see text).

the available data up to 45 T is well described by the WHH (Werthamer-Helfand-Hohenberg) model [5] for the orbital limited upper critical field. Whereas for the AD sample, the WHH model which predicts

$$B_{c2}^*(0) = 0.69\ T_c\ (\mathrm{d}B_{c2}/\mathrm{d}T)_{Tc} = 106\ \mathrm{T} \qquad (1)$$

at $T = 0$, fits the experimental data up to 30 T, only. For applied fields above 30 T increasing deviations from the WHH curve are clearly visible both for the $B_{c2}(T)$ data from the IFW and the FZD. The flattening of $B_{c2}(T)$ at high fields points to its limitation by the Pauli spin paramagnetism. This effect is measured in the WHH model by the Maki parameter $\alpha = \sqrt{2}\ B_{c2}^*(0)/B_p(0)$, where $B_p(0)$ is the Pauli limiting field. The paramagnetically limited upper critical field, B_{c2}^{P}, is given by

$$B_{c2}^{P}(0) = B_{c2}^*(0)\ /\ (1+\alpha^2)^{0.5} \qquad (2)$$

For our AD sample, a reasonable fit of the experimental data to this model has been obtained for α = 1.31 (see dotted line in figure 2). Using $B_{c2}^*(0)$ = 106 T, one obtains $B_{c2}^{P}(0)$ = 63 T for the paramagnetically limited upper critical field.

In contrast, $B_{c2}(T)$ data for a clean, but underdoped $LaO_{0.93}F_{0.07}FeAs$ sample [6] (see figure 3) show almost no Pauli limiting behavior (PLB) for fields up to 70 T. But a similar flattening of $B_{c2}(T)$ as we found for our AD sample has been reported for KFe_2As_2 [7], $SmO_{0.8}F_{0.2}FeAs$ [8] and $FeSe_{0.25}Te_{0.75}$ [9] or has been recognized by us [10] for data reported for $Ba(Fe_{0.9}Co_{0.1})_2As_2$ [11], $Ba_{0.55}K_{0.45}Fe_2As_2$ [12] and $NdO_{0.7}F_{0.3}FeAs$ [13] single crystals, in all cases for applied fields $H \parallel ab$. This is shown in figure 3 using normalized dimensionless units $b^* = B_{c2}(t)/\ [T_c\ (\mathrm{d}B_{c2}/\mathrm{d}T)_{Tc}]$ and $t = T/T_c$. The plot of b^* vs. t in figure 3 illustrates the PLB because the experimental $B_{c2}(T)$ data are normalized with respect to the orbital limited $B_{c2}^*(T)$ (see equation (1)). According to our analysis, the reported data in figure 3 are well described by the WHH model using the Maki parameters α as shown. The deviation of $b^*(t)$ at low T from the orbital $b^*(t)$ for α = 0 increases with α due to rising paramagnetic breaking. Thus, PLB or its onset is found in a rather large number of ferro-pnictide superconductors.

3.2. Vortex matter phase diagram

The upper critical field data $B_{c2}^{ab}(T)$ obtained from resistance measurements is related to a certain

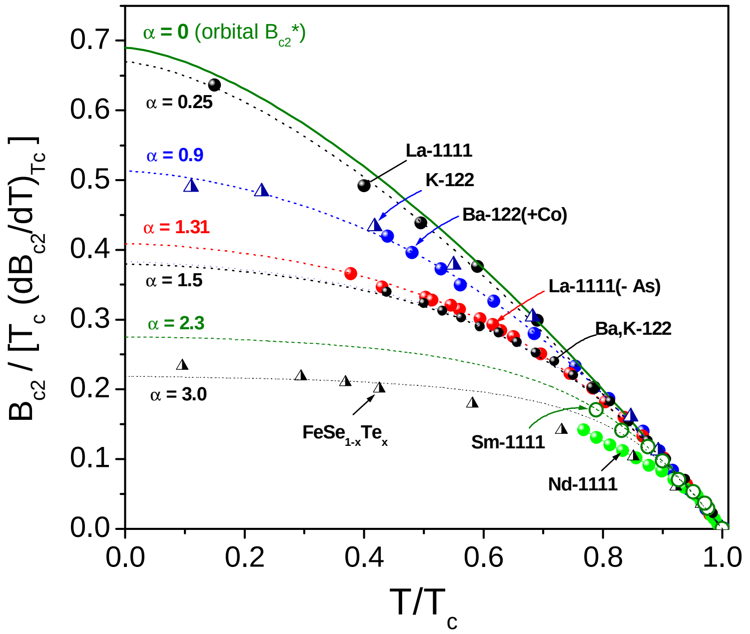

Figure 3. Normalized upper critical field b^* vs T/T_c for an AD sample (La-1111(-As)) in comparison with data reported for $LaO_{0.93}F_{0.07}FeAs$ (La-1111, T_c = 25 K) [6], KFe_2As_2 (K-122, T_c = 2.8 K) [7,14], $SmO_{0.8}F_{0.2}FeAs$ (Sm-1111, T_c = 41 K) [8], $FeSe_{0.25}Te_{0.75}$ (T_c = 14.1 K) [9,14] $Ba(Fe_{0.9}Co_{0.1})_2As_2$ (Ba-122(+Co), T_c = 21.9 K) [11], $Ba_{0.55}K_{0.45}Fe_2As_2$ (Ba-122, T_c = 32 K) [12] and $NdO_{0.7}F_{0.3}FeAs$ (Nd-1111, T_c = 45.6 K) [13]. Dotted and solid lines: WHH model for the indicated α values. All curves correspond to $H \parallel ab$.

percolative current path through the polycrystalline sample. We also determined the upper critical field $B_{c2}(T)$ by means of magnetization measurements. In figure 4 (left), $m(T)$ data for the AD sample are shown at an applied field of 6T for the zero-field cooling (ZFC) and the field cooling (FC) mode. In order to evaluate the highest and lowest upper critical fields of the polycrystalline samples, $B_{c2,max}^{ab}(T)$ and $B_{c2,min}^{c}(T)$, respectively, we used a method proposed recently by Bud'ko *et al.*[15]. According to this method, the grains with the lowest and highest $B_{c2}(T)$ can be detected by two characteristic kinks in dm/dT appearing at the temperatures T_{low} and T_{high} within the reversible part of the $m(T)$ curve (see figure 4b). Furthermore, the irreversibility field B_{irr} was defined at the temperature T_{irr} of the transition from the irreversible to the reversible part of the $m(T)$ curve.

In figure 5, the obtained vortex matter phase diagram of the AD sample and the clean reference sample are compared. We notice that in both cases, the upper critical fields $B_{c2,max}^{ab}$ and $B_{c2,min}^{c}$ are

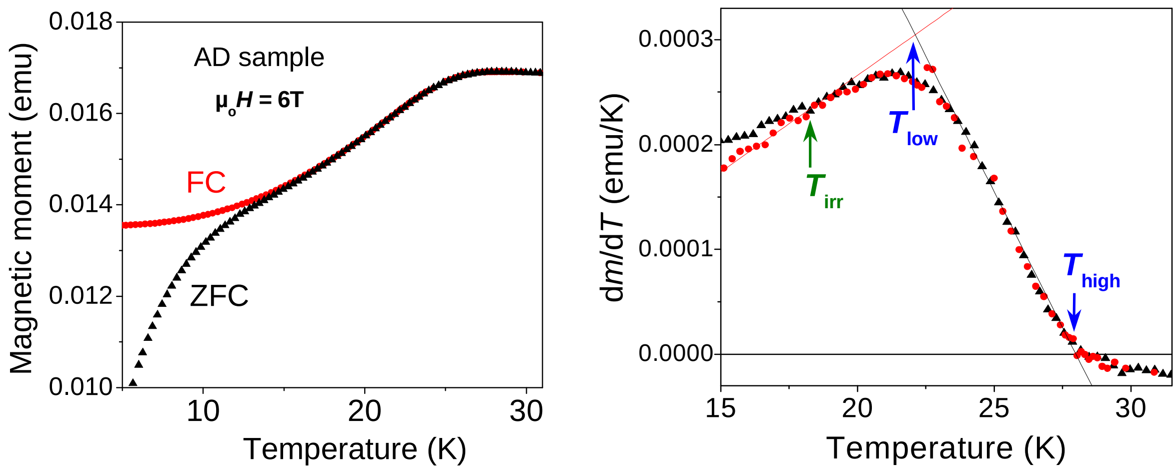

Figure 4. Temperature dependence of the magnetic moment $m(T)$ (left) and of dm/dT (right) for the AD sample measured at 6T applied after zero-field cooling (ZFC) and before cooling (FC).

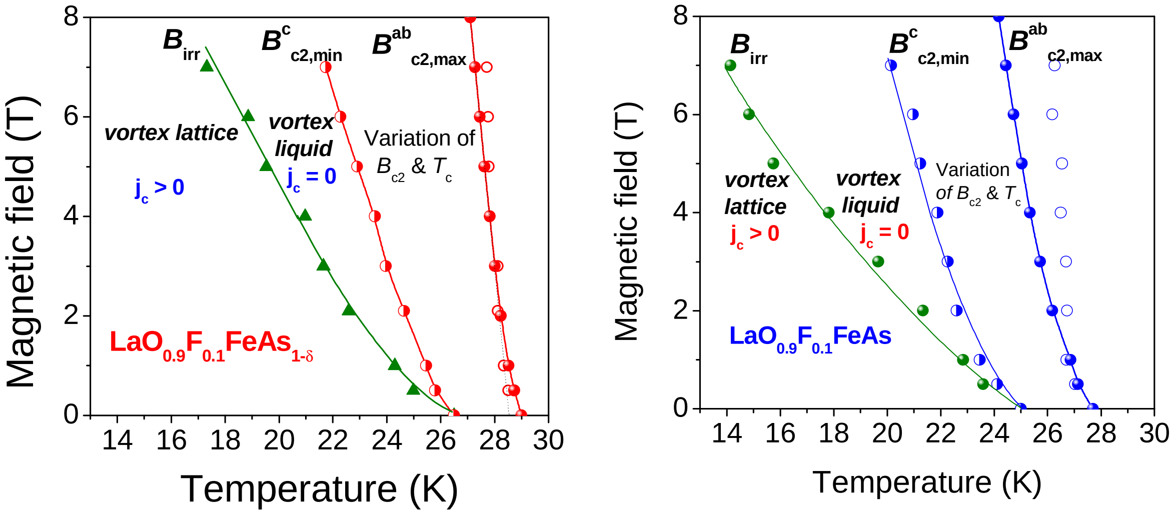

Figure 5. Temperature dependence of the highest and the lowest upper critical field $B_{c2,mag}^{ab}$ and $B_{c2,min}^{c}$, respectively, and of the irreversibility field B_{irr} for the AD sample (left) and the clean reference sample (right). In the case of $B_{c2,mag}^{ab}$, data from magnetization (open circles) and from resistance measurements (filled circles) are compared.

related to different transition temperatures T_c^{max} and T_c^{min}, respectively. Thus, the large difference between both $B_{c2}(T)$ curves is not only due to the anisotropy of the upper critical field, but also due to a significant variation of T_c within the polycrystalline samples which is found to be in the range of 10% in both samples. Obviously, the irreversibility line $B_{irr}(T)$ is related to the lower T_c^{min} as $B_{c2,min}^{c}(T)$.

The comparison of $B_{c2,mag}^{ab}$ data derived from $M(T,H)$ and $R(T,H)$ data shows a remarkably good agreement for the AD sample. In contrast, a significant difference between both data sets arises at high applied fields for the reference sample.

Furthermore, the vortex matter phase diagram of the AD sample is shifted to higher temperatures than that of the reference sample which is mainly due to the higher transition temperatures of the grains in the AD sample varying between $T_c^{min} \approx 26.5$ K and $T_c^{max} \approx 29.0$ K compared to $T_c^{min} \approx 25.0$ K and $T_c^{max} \approx 27.7$ K for the reference sample. In addition, the range of the vortex liquid phase becomes narrower in the AD sample than in the clean reference sample.

3.3. Critical current density

In FeAs based superconductors, global or intergranular currents are either negligible or much smaller than local or intragranular currents. Both current densities can be estimated by measuring the remnant magnetization as a function of the applied magnetic field. In this way, we confirmed the existence of inter- and intra- granular currents in the clean reference sample. In contrast, the intergranular currents in the AD sample are very small and could be detected only by transport measurements. We will focus here on the results for the AD sample, whereas the data for the reference sample will be presented elsewhere [16].

The field dependence of the intragrain current density j_c^{grain} in the AD sample shown in figure 6 (left) was estimated from the expression $j_c^{grain} = 30\ \Delta M/d$, where ΔM (in emu/cm^3) is the width of hysteresis loops and d (in cm) is the diameter of the grains. According to SEM images, the average grain size in this sample is roughly given by $d \approx 1\mu$m. Whereas the intragrain current density is in the range of 1MA/cm^2, the transport current in the AD sample due to percolative supercurrents flowing across some intergranular paths is very small as shown in figure 6 (right).

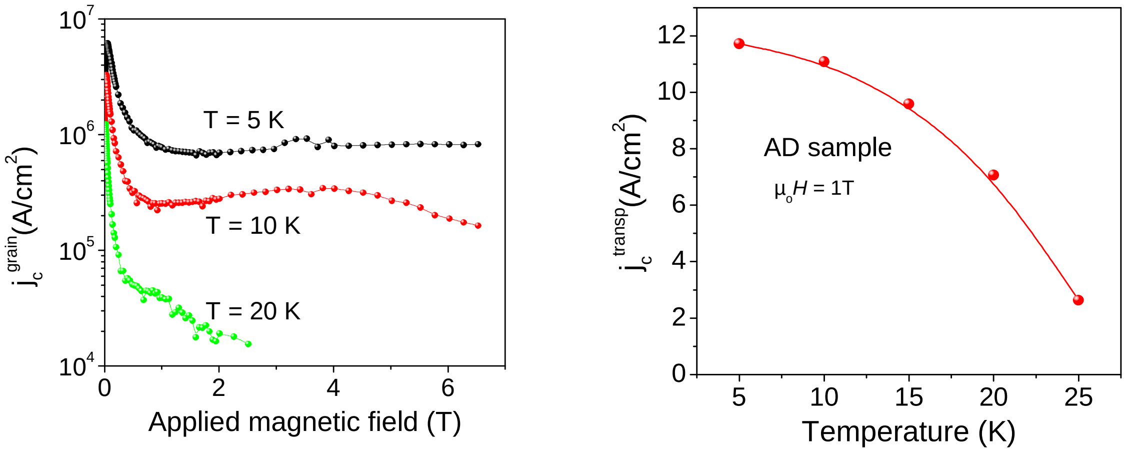

Figure 6. Field dependence of the intragrain current density of the AD sample at three temperatures (left) and temperature dependence of the transport current at an applied field of 1T (right).

4. Conclusions

Arsenic deficient $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$ samples were found to exhibit enhanced values of T_c and dB_{c2}/dT near T_c, but also a pronounced flattening at fields above 30 T which evidences PLB. We found similar effects also in published data for other FeAs based superconductors at fields H||ab. Meanwhile, in total at least seven different ferro-pnictide superconductors show PLB which seems to become a rather general feature. We found from magnetization measurements a similar variation of the transition temperature between T_c^{min} and T_c^{max} (with $\Delta T_c = T_c^{max} - T_c^{min} \approx 2.5$ K) in the AD and the clean reference $LaO_{0.9}F_{0.1}FeAs$ samples. However, the transition temperatures of the grains in the AD sample are enhanced compared to those of the reference sample. Furthermore, a more extended vortex lattice range is found for the AD sample which benefits from a rather steep irreversibility line. This is in line with a strong intragrain pinning effect observed for the AD samples, which contrasts with the very small intergranular current density found for these samples.

5. References

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