

Upper critical field, Hall effect and magnetoresistance in the iron-based layered superconductor $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$

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Abstract

By using a two-step method, we successfully synthesized the new iron-based superconductor $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$. The resistive transition curves under different magnetic fields were measured, leading to the determination of the upper critical field $H_{c2}(T)$ of this new superconductor. The value of H_{c2} at zero temperature is estimated to be about 50 T roughly. In addition, the Hall effect and magnetoresistance were measured in a wide temperature region. A negative Hall coefficient R_H has been found, implying a dominant conduction mainly by electron-like charge carriers in this material. The charge carrier density determined at 100 K is about $9.8 \times 10^{20} \text{ cm}^{-3}$, which is close to the cuprate superconductors. It is further found that the magnetoresistance does not follow Kohler's law. Meanwhile, the different temperature-dependent behaviors of resistivity, Hall coefficient and magnetoresistance have anomalous properties at about 230 K, which may be induced by some exotic scattering mechanism.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The recently discovered [1–7] iron-based superconducting oxides with a transition temperature as high as 55 K has attracted much attention. As long as a new superconductor is being discovered, the superconducting mechanism urgently needs to be understood and the transition temperature is enhanced hopefully. Thus the fundamental parameters, such as the upper critical field H_{c2} , charge carrier density and its type, the electron scattering mechanism, etc, are very important in identifying the superconducting mechanism. It is well known that the normal state properties, such as the Hall effect and magnetoresistance, are very important in learning about the electronic scattering feature, which is intimately related to the mechanism of a superconductor. For example, the superlinear temperature dependence of the normal state resistivity and the clear temperature dependence of the Hall coefficient in a wide temperature region in cuprate superconductors have been considered as two important anomalous properties which

suggest an unusual scattering process beyond electron–phonon scattering. In this paper we report the successful fabrication of this material by a two-step method, and the measurements on the resistive transition under different magnetic fields, Hall effect and magnetoresistance. By analyzing the data we obtained fresh information about the upper critical field, the charge carrier density and its type, as well as the scattering times, etc.

2. Sample preparation and characterization

The polycrystalline samples were synthesized by using a two-step solid state reaction method. First the starting materials Fe powder (purity 99.95%) and As grains (purity 99.99%) were mixed in 1:1 ratio, ground and pressed into a pellet shape. Then it was sealed in an evacuated quartz tube, followed by heat treating at 700 °C for 10 h. The resultant pellet was crushed and ground together with LaF_3 powder (purity 99.95%), La_2O_3 powder (purity 99.9%) and grains of La (purity 99.99%) in

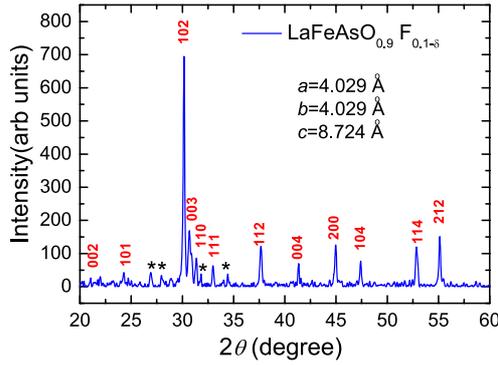


Figure 1. X-ray diffraction patterns for the sample $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$. Almost all main peaks can be indexed by a tetragonal structure with $a = b = 4.029 \text{ \AA}$ and $c = 8.724 \text{ \AA}$. The tiny peaks marked by asterisks are from the impurity phase, perhaps FeAs.

stoichiometric amounts as in the formula $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$. Again it was pressed into a pellet and sealed in an evacuated quartz tube and heated at about 940°C for 2 h, followed by heat treating at 1150°C for 48 h. Then it was cooled down slowly to room temperature. Since a little amount of F may escape during the second step, in the formula for our sample, we use $0.1 - \delta$ as the possible concentration of F. In figure 1, we show the x-ray diffraction patterns for the sample $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$. Almost all main peaks can be indexed by a tetragonal structure with $a = b = 4.029 \text{ \AA}$ and $c = 8.724 \text{ \AA}$. Therefore the dominant component is from $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$ with only tiny peaks at the same scale as appeared in the data of the original paper [1]. This tiny amount of second phase, perhaps from FeAs, together with the granular behavior of the samples may have some influence on the zero resistance point, but they should not have any obvious influence on the upper critical field and the normal state properties.

3. Experimental data and discussion

The resistance and Hall effect measurements were done in a physical properties measurement system (Quantum Design, PPMS) with a magnetic field up to 9 T. The six-lead method was used in the measurement on the longitudinal and the transverse resistivity at the same time. The resistance was measured by either sweeping the magnetic field at a fixed temperature or sweeping the temperature in a fixed field. The temperature stabilization was better than 0.1% and the resolution of the voltmeter was better than 10 nV.

3.1. Resistive transition and upper critical field

In figure 2 we present the temperature dependence of resistivity for the $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$ sample under different magnetic fields. One can see that the onset transition point shifts weakly with the magnetic field, but the zero resistance point shifts more quickly to lower temperatures. This is understandable since the latter is determined by the weak links between the grains as well as the vortex flow behavior, while the former is controlled by the upper critical field of the individual grains. This allows us to determine the upper critical field of this

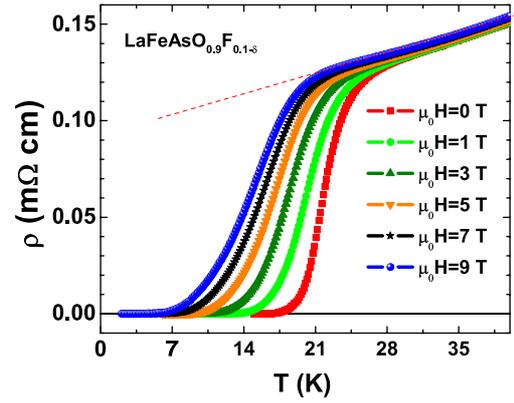


Figure 2. Temperature dependence of resistivity for the $\text{LaFeAsO}_{0.9}\text{F}_{0.1-\delta}$ bulk sample under different magnetic fields. The onset transition point defined by 99% ρ_n shifts weakly with the magnetic field. The dashed line indicates the extrapolation of the normal state resistivity.

material. However, we should mention that the upper critical field determined in this way reflects mainly the situation of the $H \parallel ab$ plane since the Cooper pairs within the grains with this configuration will last to the highest field compared to other grains. Taking the onset point of the transition as the upper critical field point $T_c(H_{c2})$ means that almost all Cooper pairs are broken at this temperature and magnetic field. By taking a criterion of 99% ρ_n (referenced by the normal state resistivity ρ_n marked by the dashed line in figure 3) we can determine the upper critical field $H_{c2}(T)$ as shown in figure 3. It is seen that the slope of $H_{c2}(T)$ near T_c , i.e. dH_{c2}/dT , is about -2.3 T K^{-1} . By using the Werthamer–Helfand–Hohenberg (WHH) formula [8] the value of the zero-temperature upper critical field $H_{c2}^{ab}(0)$ can be estimated by

$$H_{c2}(0) = -0.693T_c \left(\frac{dH_{c2}}{dT} \right)_{T=T_c}. \quad (1)$$

Taking $T_c = 28.9 \text{ K}$, we get $H_{c2}(0) \approx 45.8 \text{ T}$ roughly. We can also determine the H_{c2} by using the formula based on the Ginzburg–Landau (GL) equation. In GL theory, it is known that $H_{c2} = \Phi_0/2\pi\xi^2$ and $\xi \propto \sqrt{(1+t^2)/(1-t^2)}$, with Φ_0 the flux quanta, ξ the coherence length and $t = T/T_c$ the reduced temperature. Thus one has

$$H_{c2}(T) = H_{c2}(0) \frac{1-t^2}{1+t^2}. \quad (2)$$

Although GL theory is especially applicable near T_c , the above equation has been proved to be satisfied in a much wider temperature regime [9]. We thus use the above equation to fit our data and show them as the solid line in figure 3. The zero-temperature upper critical field $H_{c2}(0)$ determined in this way is $H_{c2}(0) = 56 \text{ T}$. This value is a bit higher than that obtained using the WHH formula. We note that a recent result reported that the WHH approximation could not be simply applied in this material [10], and even $H_{c2}(0)$ is affected by the multiband property [11]. However, our result can give a rough magnitude of $H_{c2}(0)$ because of the limit of the magnetic field.

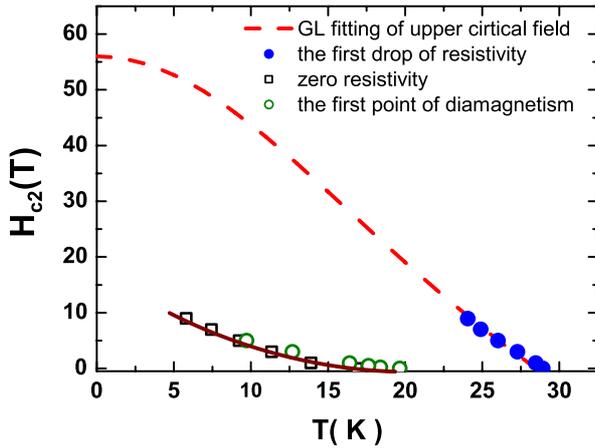


Figure 3. Phase diagram derived from the resistive transition curves. The onset transition point gives rise to the upper critical field H_{c2}^{ab} shown by the filled circles. The dashed line shows the theoretical curve based on GL theory (equation (2)). The magnetic onset transition point and the zero resistivity point are quite close to each other, which are shown by the open circles and squares, respectively.

3.2. Magnetoresistance

In figure 4 we show the resistance transition of the sample at zero field in a wide temperature region by open circles. From that we can get the zero resistance temperature at about 19 K and the onset temperature 28.9 K (99% of the normal state resistivity). The resistivity at 30 K is 0.133 m Ω cm, while the residual resistance ratio $RRR \equiv \rho(300 \text{ K})/\rho(30 \text{ K})$ is about 18.1. This may indicate the good quality of the sample in our experiments. The general shape of the resistivity curve at this doping shows a very good metallic behavior. The inset shown in figure 4 gives the zero-field-cooled and also the field-cooled DC magnetization of the sample at 20 Oe. The onset critical temperature by magnetic measurements is about 24 K (see from an enlarged view), which corresponds to the middle transition point of resistance.

Magnetoresistance is a very powerful tool to investigate the electronic scattering process and the information about the Fermi surface. For example, in MgB₂, a large magnetoresistance (MR) was found which is closely related to the multiband property [12, 13]. For this new superconductor, we also measured and found a clear MR. In figure 5(a) we present the field dependence of the MR ratio, i.e. $\Delta\rho/\rho_0$, where ρ is the resistivity, ρ_0 is the resistivity at zero field and $\Delta\rho = \rho(H) - \rho(0)$. One can see that the MR is about 2.2% at 40 K and 9 T for this sample. This ratio is one order of magnitude smaller than MgB₂ with the same RRR. However, considering that the sample is a polycrystalline sample, the MR effect may be weakened by mixing the transport components with the magnetic field along different directions of the crystallographic axes. For a single band metal with a symmetric Fermi surface, Kohler's law is normally obeyed. Kohler's law [14] shows that the magnetoresistance $\Delta\rho/\rho_0$ measured at different temperatures should be scalable with the variable H/ρ_0 . For MgB₂, Kohler's law is not obeyed because of the multiband property [12]. We also do scaling based on Kohler's law for this sample: the result is shown in figure 5(b). Clearly, the data

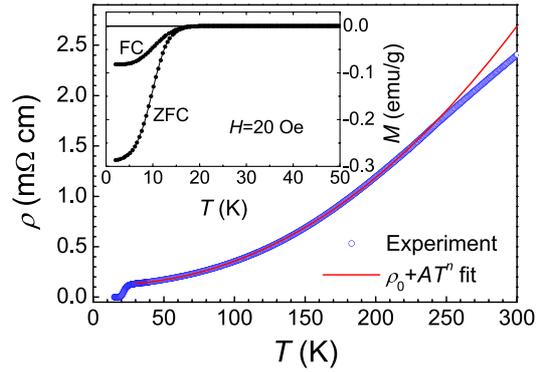


Figure 4. Temperature dependence of resistivity for the LaFeAsO_{0.9}F_{0.1- δ} bulk sample (shown by symbols) and the solid line shows the fit in the low temperature range by $\rho_0 + AT^n$ with $\rho_0 = 0.117 \text{ m}\Omega \text{ cm}$ and $n = 2.16$. Above about 220 K, the fitting curve deviates clearly from the data. The inset shows the temperature dependence of DC magnetization for the zero-field cooling (ZFC) and field cooling (FC) at $H = 20 \text{ Oe}$.

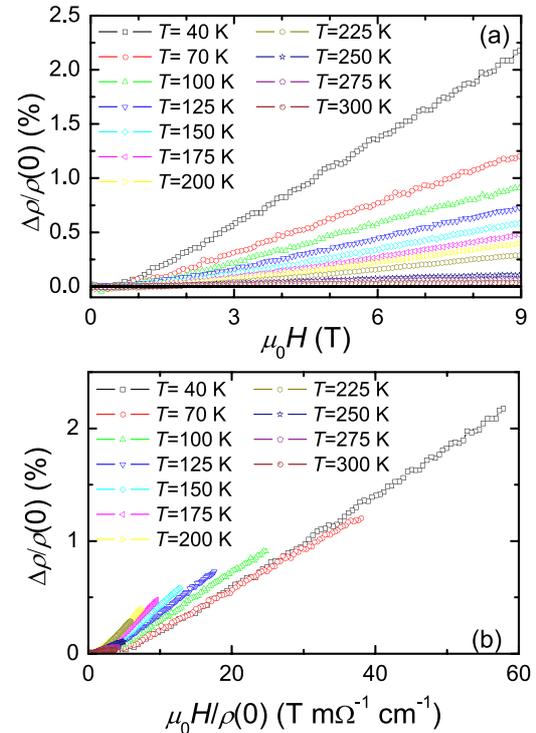


Figure 5. (a) Field dependence of magnetoresistance $\Delta\rho/\rho(0)$ at different temperatures. One can see that the MR decreases rapidly when the temperature is increased. Above about 250 K, the MR becomes very weak. (b) Kohler plot for the sample at different temperatures, and Kohler's rule is not obeyed.

measured at different temperatures do not overlap and Kohler's law is violated for this material. This discrepancy may suggest that in this new material there may be a multiband effect or other exotic scattering, such as spin scattering, which depends on the magnetic field. However, the MR in this sample does show something special. For two-band or multiband materials with weak MR, the MR ratios could be well described by the

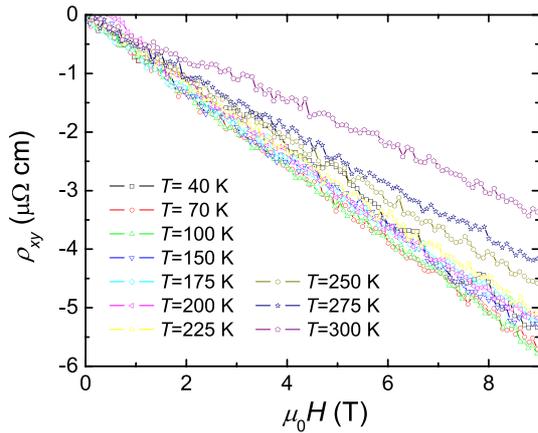


Figure 6. ρ_{xy} versus the magnetic field $\mu_0 H$ at different temperatures. The curves at temperatures below 250 K have similar behaviors, while at temperatures above that temperature, the absolute values of slopes have a sudden decrease.

expression $\Delta\rho/\rho_0 \propto H^2$ in the low field region. This is because the contribution of the higher-order even terms of $\mu_0 H$ could be omitted at a low field (the odd terms are absent here according to the Boltzmann equation for electronic transport). This effect is also found in NbSe₂ which has a complex Fermi surface structure [15]. It was reported that both LaFeAsO [16] and LaFePO [17] have five orbitals crossing the Fermi level, and Fermi surfaces with both electron and hole type band are present. In this sense, the anomalies mentioned above in the new superconductor may also be induced by a multiband effect together with complex Fermi surfaces. In single-crystal samples, when the field is along the *c* axis, a stronger in-plane magnetoresistance is expected.

3.3. Hall effect

For a normal metal with Fermi liquid feature, the Hall coefficient is a constant versus temperature. However, this situation is changed for a multiband material or a sample with non-Fermi liquid behavior, such as the cuprate superconductors. We also take the Hall effect measurement for this sample. As shown in figure 6, the transverse resistivity remains negative at all temperatures above the critical temperature, indicating that the electric transport is dominated by electron-like charge carriers, not hole-like ones. For a clean sample, the nonlinear Hall effect is also a sign of a multiband, and the effect is weaker in dirtier samples [13]. In our measurements, all curves shown in figure 6 have good linearity versus the magnetic field, which may be caused by the disorders within the sample. From this set of data, the Hall coefficient $R_H = \rho_{xy}/H$ is determined and shown in figure 7(a). It is clear that R_H is almost a constant below about 250 K, and there is a kink at about 150 K considering the small error shown in the figure. The charge carrier density calculated by $n = 1/R_H e$ is about $9.8 \times 10^{20} \text{ cm}^{-3}$ which is very close to the cuprate superconductors. This may imply that the superfluid density in this superconductor is also very diluted, as suggested by a recent theoretical

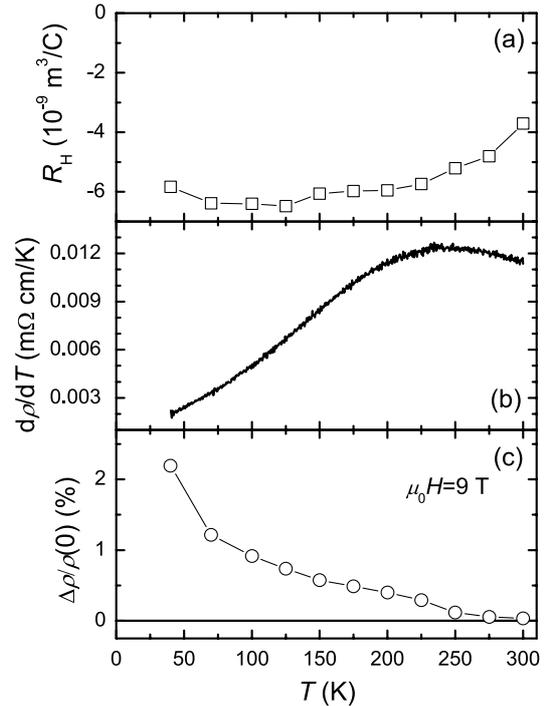


Figure 7. Temperature dependence of the Hall coefficient (a), differential of the ρ - T curve (b) and the magnetoresistance ratio at 9 T (c). There are obvious turning points at about 230 for both of the curves shown in (a) and (b), while the magnetoresistance shows a sudden decrease at that point.

proposal [16]. However, it should be noted that the Hall coefficient R_H could not be simply expressed as $1/ne$ for a multiband material [13], so it needs further consideration if a multiband property dominates the electric transport. From figure 6, the transverse resistivity curves almost overlap at $T < 250$ K. However, at a temperature above 250 K, the absolute value of the slope, i.e. the Hall coefficient R_H , decreases rapidly. As shown in figure 7(a), R_H behaves a little different from another recent work on LaFeAsO_{0.9}F_{1- δ} [18]. In our sample, R_H shows a weak T dependence at the temperature below 225 K, while at $T > 250$ K, the absolute value of R_H has a rapid decrease. This is similar to the situation of MgCNi₃ [19], which may be caused by the exotic scattering when a ferromagnetic fluctuation is present. The temperature-dependent Hall coefficient also tells us that either the multiband effect or some other unusual scattering process may be involved in the electron conduction in the material.

In order to reveal an anomalous behavior at about 230 K, here we give a further discussion. The differential of the ρ - T curve shown in figure 4 is given in figure 7(b). It is clear that there is a maximum value at about 240 K of the differential slope $d\rho/dT$. This is similar to the situation reported in the underdoped LaOFeAs sample [1], where they found a clear kink point of resistivity and marked the temperature point as T_{anom} . In our present sample, this kink is absent but the resistivity shows a weak downward feature around 240 K, as evidenced by a bump on the curve of $d\rho/dT$ versus T . Therefore the anomaly at about 240 K in our sample may be related to the situation at T_{anom} in the original paper [1].

We also try to fit the normal state ρ - T curve with the formula $\rho_0 + AT^n$. As shown in figure 4, the curve below 240 K could be well fitted and the fitting results give values of $n = 2.16$ and $\rho_0 = 0.117 \text{ m}\Omega \text{ cm}$. However, just from this temperature and above, the fitting curve starts deviating from the data. As shown in figure 7(c), there is also a sudden decrease of MR at a temperature of 240 K. The origin of this anomaly at about 230 K certainly needs further consideration.

So far, a weak magnetic signal has been measured in the sample in the normal state, which is exactly the same as that reported in the original paper by Kamihara *et al* [1]. Magnetic measurements show that this magnetic signal even has a small hysteresis in the low field region. At this moment, we do not know whether this magnetic signal is due to an intrinsic feature of the LaFeAsO phase, or is due to the second tiny impure phase. If the former is right, some exotic scattering, like the electron-magnon scattering or magnetic skew scattering, would exist. This deserves further study on improved samples. Our present investigation will thus provide a basic platform for future studies.

4. Conclusions

In summary, the temperature dependence of resistivity under different magnetic fields, the magnetoresistance and Hall coefficient have been measured in the newly found layered superconductor LaFeAsO_{0.9}F_{0.1- δ} . The value of H_{c2} at zero temperature is obtained roughly to be about 50 T. The Hall coefficient is negative, indicating that the electron-like charge carriers dominate the electrical transport. The charge carrier density at 100 K is about $9.8 \times 10^{20} \text{ cm}^{-3}$ showing that the superconductor may have a diluted superfluid, as in cuprate superconductors. The Hall coefficient R_H has a weak temperature dependence below 230 K, but it rises more rapidly above that temperature. At the similar temperature the magnetoresistance becomes very small, together with a maximum of the differential of the resistivity curve $\rho(T)$. The $\rho(T)$ curve at low temperature could be fitted by $\rho_0 + AT^n$ with $n = 2.16$. Kohler's law is clearly violated in all temperature regions. These observations can be explained by the multiband effect or

some exotic scattering, like the scattering with magnetic moments or in the presence of weak magnetic correlation.

Acknowledgments

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