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Superconductivity at 36 K in gadolinium-arsenide oxides $GdO_{1-x}F_xFeAs$*

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In this paper we report the fabrication and superconducting properties of $GdO_{1-x}F_xFeAs$. It was found that when x is equal to 0.17, $GdO_{0.83}F_{0.17}FeAs$ is a superconductor with the onset transition temperature $T_c^{on} \approx 36.6$ K. Resistivity anomaly near 130 K was observed for all samples up to $x = 0.17$, and such a phenomenon is similar to that of $LaO_{1-x}F_xFeAs$. Hall coefficient indicates that $GdO_{1-x}F_xFeAs$ is conducted by electron-like charge carriers.

iron-based superconductor, high-temperature superconductor

1 Introduction

Since the discovery of superconductivity in iron-based layered quaternary compound LaOFeP[1], extensive efforts have been devoted to searching for new superconductors among this system[1−10]. It was found that with the replacement of P by As and partial substitution of O with F, $LaO_{1-x}F_xFeAs$ changes into the superconducting state below $T_c \approx 26$ K[2]. Subsequently superconductivity at 25 K was also observed in $La_{1-x}Sr_xOFeAs$ in which no F was added into the sample, therefore it was a hole-doped superconductor[3]. More recently, superconductors $LnO_{1-x}F_xFeAs$ with light rare-earth substitution (Ln = Ce, Pr, Sm) were realized and superconducting transition temperature T_c was raised to 52 K[4−7]. As to the heavy rare-earth element, however, single phase could not be easily formed and no superconducting state was observed below 2 K. As to the element Gadolinium which locates near the heavy rare-earth element, experimentally a drop of resistivity was observed below 10 K but with a residual resistivity down to 2 K[8]. So it is worth exploring further that whether $GdO_{1-x}F_xFeAs$ is also a superconductor with much higher T_c. In this study, we report the superconducting properties of $GdO_{0.83}F_{0.17}FeAs$ with the onset transition

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temperature T_c^{on} of about 36.6 K.

2 Experiment

Polycrystalline samples $GdO_{1-x}F_xFeAs$ (x = 0.12, 0.15, 0.17) were synthesized by a conventional solid state sintering method. The raw materials are all high purity Gd_2O_3 (99.99%), GdF_3 (99.99%), Fe (99.95%), As (99.99%), and Gd (99.99%). The detailed synthesis method is the same as that in the papers we reported previously[9,10]. The as-sintered pellet is concrete ceramic-like with dark-brown surface. X-ray diffraction measurement was performed at room temperature using an MXP18A-HF-type diffractometer with Cu-K_α radiation from 10° to 80° with a step of 0.01°. The magnetization measurements were carried out on a Quantum Design superconducting quantum interference device (SQUID) magnetometer. The electrical resistivity and Hall coefficient were measured by a Physical Property Measurement System (PPMS, Quantum Design) with a standard six-probe method.

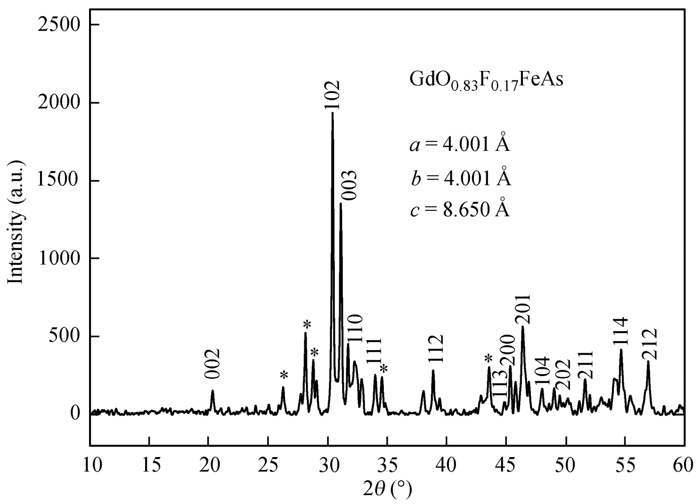

Figure 1 XRD pattern of $GdO_{1-x}F_xFeAs$ (x = 0.17). It is clear that the dominant phase is $GdO_{1-x}F_xFeAs$. The asterisks mark the peaks from the impurity phase.

Figure 1 shows the X-ray diffraction (XRD) pattern of the sample $GdO_{0.83}F_{0.17}FeAs$. The pattern can be indexed in the tetragonal space group with $a = b$ = 4.001 Å and c = 8.650 Å. Obviously the phase is dominated by $GdO_{1-x}F_xFeAs$, though minor impurity phases still exist as marked by the asterisks. Such impurity phases could be caused by the inadequate sintering temperature 1160℃ in our experiment. The indices of the crystal lattice we obtained are consistent with the counterparts of $LnO_{1-x}F_xFeAs$ (Ln = Ce, Pr, Sm)[4−7].

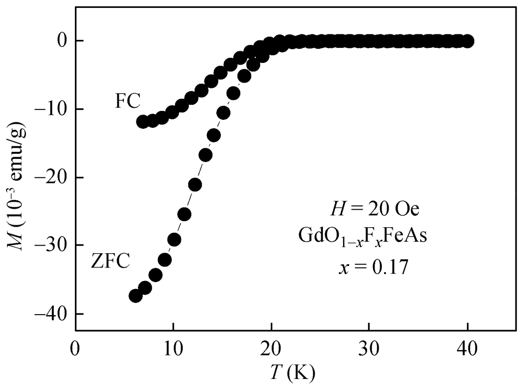

Figure 2 DC magnetization of $GdO_{0.83}F_{0.17}FeAs$ measured in the zero-field-cooled (ZFC) and field-cooled (FC) processes. A diamagnetic signal is easily observed at about 22 K.

Figure 2 shows the temperature dependence of DC magnetization for sample $GdO_{0.83}F_{0.17}FeAs$. The diamagnetic signal appears below 22 K, and a simple estimation on the magnetization at 2 K reveals that the superconducting volume fraction is more than 40%. It should be noted that the response to the magnetic field in the normal state of $GdO_{0.83}F_{0.17}FeAs$ is paramagnetic with a minor value compared to the diamagnetic value and such a contribution to magnetization is subtracted as a background (the same to ZFC and FC curve).

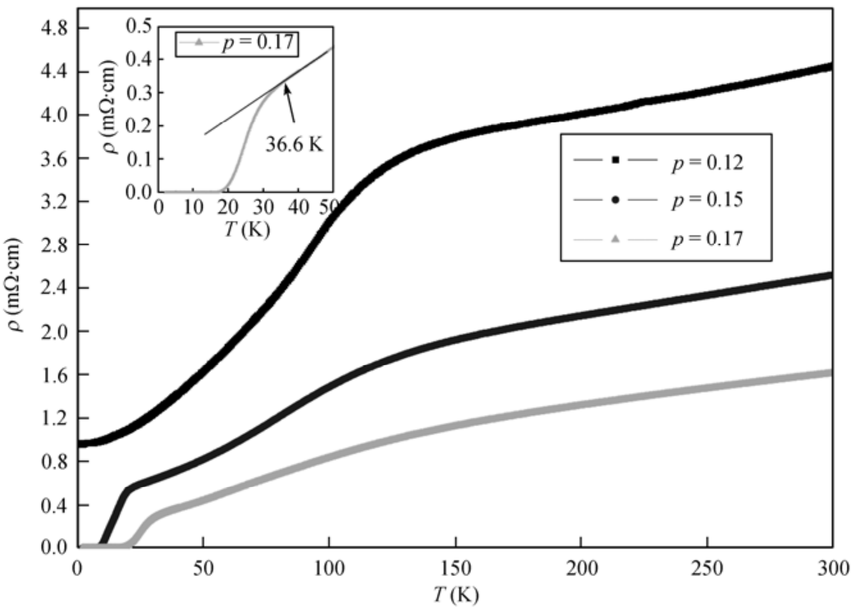

Figure 3 The electrical resistivity vs. temperature for $GdO_{1-x}F_xFeAs$ (x = 0.12,0.15,0.17). The inset shows the enlarged view of the superconducting transition area for the sample x = 0.17. The onset transition temperature is defined at the point where the resistivity starts to deviate from the normal state background as marked by the straight line here.

The temperature dependence of resistivity of all three samples is shown in Figure 3. We can see that the resistivity shows an anomaly around 130 K, and this anomaly weakens with more F doping. Such an anomaly and corresponding evolution with F doping have been observed in other Fe-based Arsenic compounds, however, the anomaly did not happen in the Nickel based Arsenic system. The obvious disparity between Fe-based and Ni-based systems deserves to be further studied. Both x = 0.15 and x = 0.17 samples exhibit superconducting transitions and zero-resistance at a lower temperature. From the inset of Figure 3 we can see the onset drop of resistivity at about 36.6 K for x = 0.17 sample. As to $GdO_{0.83}F_{0.17}FeAs$, a slight hump near 130 K was also observed in the resistivity curve, which suggests that T_c could be increased further as long as more Fluorines were doped into $GdO_{1-x}F_xFeAs$. The transition width can also be narrowed in a refined fabrication process in the future.

Hall effect measurement for sample $GdO_{0.83}F_{0.17}FeAs$ is shown in Figure 4. The transverse resistivities ρ_{xy} above T_c are all negative, indicating that the normal state conduction of $GdO_{0.83}F_{0.17}FeAs$ is dominated by the electron-like charge carriers. The Hall coefficient $R_H = \rho_{xy}$ changes slightly at high temperatures but drops below 100 K. The value of R_H is about -1×10^{-8} m^3/C at 100 K, and compared with that of $LaO_{0.9}F_{0.1}FeAs$, the value of Hall coefficient is similar[9]. An estimation based on the single band model gives a charge carrier density of 1×10^{21} cm^{-3}.

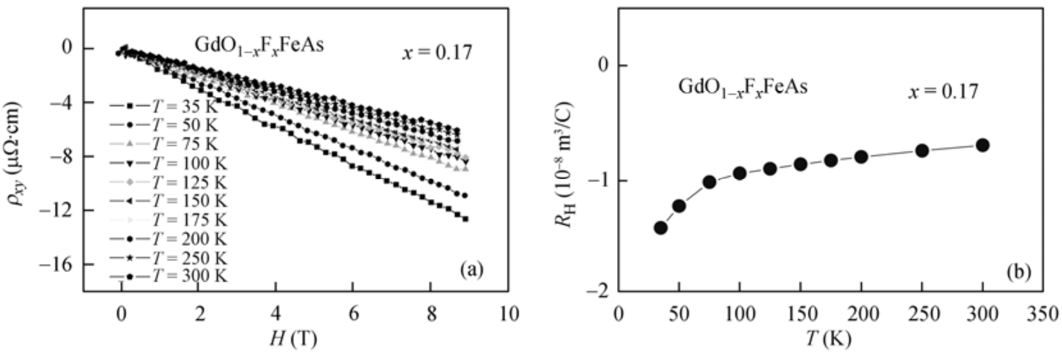

Figure 4 (a) Transverse resistivity vs. magnetic field at different temperatures for $GdO_{0.83}F_{0.17}FeAs$; (b) temperature dependence of Hall coefficient for $GdO_{0.83}F_{0.17}FeAs$. The negative value indicates that the charge carrier is of the electron type.

3 Conclusions

In this study we report the fabrication and the superconducting properties of $GdO_{1-x}F_xFeAs$. It was found that as x is equal to 0.17, $GdO_{0.83}F_{0.17}FeAs$ is a superconductor with the onset transition temperature of about 36.6 K. Resistivity anomaly near 130 K was observed for all samples, which is similar to that of $LaO_{1-x}F_xFeAs$. Hall coefficient suggests that $GdO_{0.83}F_{0.17}FeAs$ is conducted by electron-like charge carriers.

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