Hall effect and magnetoresistance in single crystals of $NdFeAsO_{1-x}F_x$ ($x=0$ and 0.18)

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Hall effect and magnetoresistance have been measured on single crystals of $NdFeAsO_{1-x}F_x$ with $x=0$ ($T_c=0$ K) and $x=0.18$ ($T_c=50$ K). For the undoped samples, strong Hall effect and magnetoresistance with strong temperature dependence were found below about 150 K. The magnetoresistance was found to be as large as 30% at 15 K at a magnetic field of 9 T. From the transport data we found that the transition near 155 K was accomplished in two steps: first one occurs at 155 K which may be associated with the structural transition, the second one takes place at about 140 K which may correspond to the spin-density-wave-like transition. In the superconducting sample with $T_c=50$ K, it is found that the Hall coefficient also reveals a strong temperature dependence with a negative sign. But the magnetoresistance becomes very weak and does not satisfy Kohler's scaling law. These dilemmatic results (strong Hall effect and very weak magnetoresistance) prevent understanding of the normal-state electric conduction by a simple multi-band model by taking into account the electron and hole pockets. Detailed analysis further indicates that the strong temperature dependence of R_H cannot be easily understood with the simple multi-band model either. A picture concerning a suppression to the density of states at the Fermi energy in lowering temperature is more reasonable. A comparison between the Hall coefficient of the undoped sample and the superconducting sample suggests that the doping may remove the nesting condition for the formation of the spin-density wave order, since both samples have very similar temperature dependence above 175 K.

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I. INTRODUCTION

Since the discovery of superconductivity at 26 K in iron-based layered quaternary compound $LaFeAsO_{1-x}F_x$,[1] a group of high-temperature superconductors have been discovered. For example, in the electron doped region the highest critical temperature $T_c=55$ K was found in $SmFeAsO_{0.9}F_{0.1}$,[2] while in the hole-doped region superconductivity at $T_c=25$ K was first found in $La_{1-x}Sr_xFeAsO$ (Ref. 3) and later $T_c=38$ K (Ref. 4) in $Ba_{1-x}K_xFe_2As_2$. At the same time extensive efforts have been devoted to study the nature of this generation of high-temperature superconductors. Among them, very high upper critical field was inferred through high-field measurements in these iron-based superconductors,[5] which indicates encouraging potential applications. Low temperature specific heat,[6] point-contact tunneling spectrum,[7] lower critical field,[8] nuclear magnetic resonance[9] (NMR), etc. revealed unconventional pairing symmetry in the superconducting state. Theoretical calculations pointed out that the superconductivity in these iron-based superconductors may emerge on several disconnected pieces of the Fermi surface,[10–14] thus exhibiting multigap effect. Many experiments have already shown that these superconductors exhibit multiband features.[5,8,15] Spin-density-wave order and structural distortion were observed in undoped LaOFeAs system.[16–18] All these experimental and theoretical works indicate an unconventional superconducting mechanism in this system. To get a deeper insight into the superconducting mechanism, it becomes very essential to know the normal-state properties. Unfortunately almost all the transport data so far in the normal state were taken from polycrystalline samples, this casts big doubts in drawing any solid conclusions. This situation becomes very serious in the layered system, such as the iron-based superconductors. Recently our group has successfully fabricated single crystals of $NdFeAsO_{1-x}F_x$ ($x=0.18$) with $T_c^{onset}=50$ K,[19] so it becomes possible to measure the intrinsic transport properties of these iron-based superconductors. Here for the first time we report the measurements of Hall effect and magnetoresistance on $NdFeAsO_{1-x}F_x$ ($x=0,0.18$) single crystals. Our data provide intrinsic and detailed information about the electron scattering in the normal state.

II. EXPERIMENT

The crystals were made by flux method using NaCl as the flux, and the detailed process of sample preparation was given elsewhere.[19] The surface of the crystal looks rather flat. X-ray diffraction pattern taken on one single crystal shows only (00l) peaks and the full-width at the half maximum (FWHM) of the (003) peak is only 0.12°, indicating good crystallinity. We made electric contacts on them using the Pt deposition method of the focused-ion-beam (FIB) technique. As shown in the insets of Fig. 1, crystals with six Pt leads are shown and the longitudinal and transverse resistance can be measured at the same time. The measurements were carried out on a physical property measurement system (PPMS) (Quantum Design) with magnetic fields perpendicular to the *ab* plane of the samples and up to 9 T. The current density during the measurement was about 400–500 A/cm^2. The in-plane longitudinal and the Hall resistance were measured by either sweeping the magnetic field at a fixed temperature or sweeping the temperature at a fixed magnetic field.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

In Fig. 1(a), the resistive transitions of two superconducting samples were shown. The superconducting (SC)

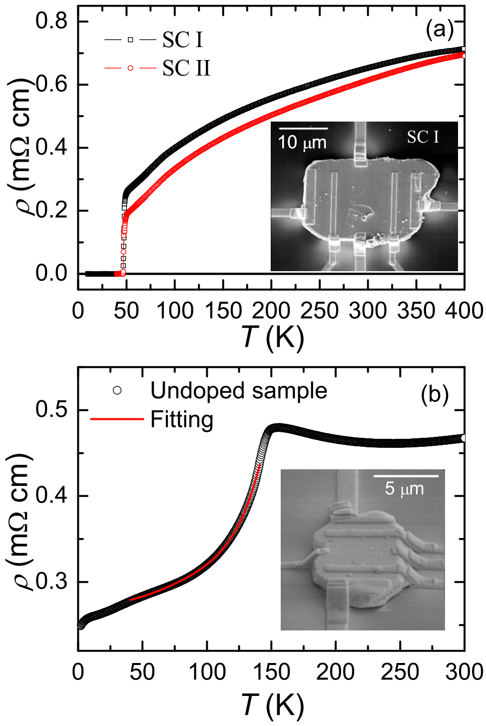

FIG. 1. (Color online) Temperature dependence of resistivity for two superconducting (SC) $NdFeAsO_{0.82}F_{0.18}$ single crystals (a) and one NdFeAsO single crystal (b). The insets show the scanning electron microscope (SEM) picture of SC sample I and undoped sample with the electric contacts of Pt metal made by focused-ion-beam technique for the resistance and Hall measurements. The solid line in (b) shows the theoretical fitting by Eq. (3) which gives the SDW energy gap about 1082 K (see text).

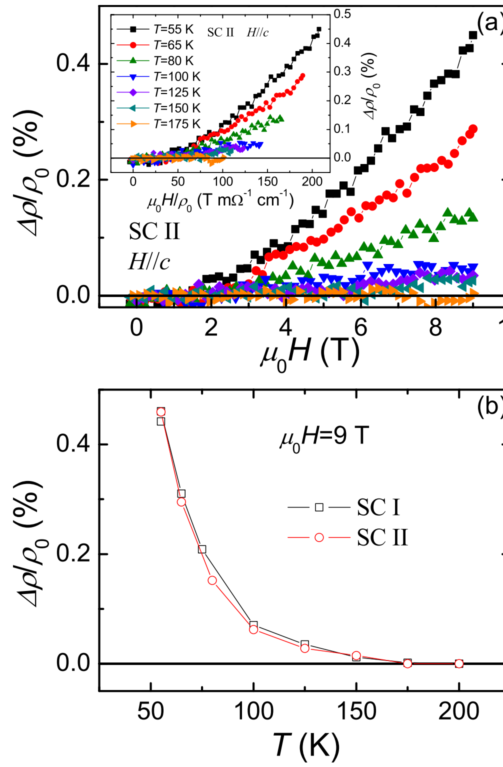

FIG. 2. (Color online) (a) Field dependence of MR $\Delta\rho/\rho_0$ for SC sample II at different temperatures. The inset shows the plot by following Kohler's law at different temperatures, one can see that Kohler's law is not obeyed. (b) Temperature dependence of MR at 9 T. One can see that the MR is rather small and decreases rapidly with increasing temperature. Above about 175 K, the MR becomes negligible in our measurement resolution.

$NdFeAsO_{0.82}F_{0.18}$ samples have very sharp transitions, i.e., the onset transition temperatures are about 50 K and the transition width is less than 2 K ($90\%\rho_n$ and $1\%\rho_n$), which indicates good quality of the samples. Obviously, the general shapes of the resistivity curves for the two samples do not show a good linear metallic behavior but a strange curved feature persists up to 400 K. The resistivity at 55 K is about 0.20 and 0.28 mΩ cm for the two samples, respectively. The slight difference between the two values may be attributed to the error in the measurement on thickness by the FIB technique, or due to the slight difference between the two samples. However, both values are much smaller than 0.58 mΩ cm in a polycrystalline sample.[20] For the resistive curve of the undoped NdFeAsO sample as shown in Fig. 1(b), a strong resistivity anomaly can be found near 150 K. This anomaly was attributed to the structural transition and/or the spin-density wave (SDW) formation. We will show below that the onset point at about 150 K of the resistive transition may be corresponding to the structural transition, and another one which occurs at a lower temperature is related to the SDW formation.[16] For Fig. 1(b), it should be noted that the unusual decrease at about 6 K could be regarded as the emergence of the ordering of the Nd ion. So the doping of F weakens the spin-density wave and the structural phase transition, and generates the superconductivity.

A. Magnetoresistance and Hall effect of superconducting $NdFeAsO_{0.82}F_{0.18}$ crystals

It is known that the magnetoresistance (MR) is a very powerful tool to investigate the electronic scattering and the message of the Fermi surface. For example, in MgB_2, a large magnetoresistance was found which is closely related to the multiband property.[21,22] Figure 2(a) shows the field dependence of MR for SC sample II. Here we define the magnetoresistivity as $\Delta\rho=\rho(H)-\rho_0$, where $\rho(H)$ is the longitudinal resistivity at a magnetic field H and ρ_0 is that at zero field. It can be observed that the MR is very weak, i.e., less than 0.5% at 55 K, which is of the same magnitude as the value in hole-doped cuprate superconductors.[23,24] Figure 2(b) shows the MR versus temperature at 9 T. One can see that MR decays rapidly with increasing temperature, and it cannot be detected in our measuring resolution above 175 K. Usually the MR effect may be weakened by mixing the transport components with the magnetic field along different directions of the crystallographic axes. However, in our measurements the MR in the single crystal is one order of magnitude smaller than the value in the polycrystalline samples.[15] For many metallic materials with a symmetric Fermi surface, Kohler's law is normally obeyed. According to Kohler's law,[25] MR at different temperatures can be scaled by the expression $\Delta\rho/\rho_0=f(H\tau)=F(H/\rho_0)$ with the assumption that

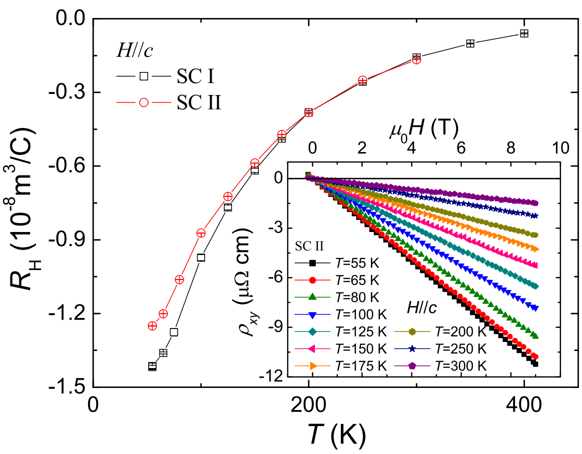

FIG. 3. (Color online) Temperature dependence of the Hall coefficient R_H for the two superconducting samples. Strong temperature dependence of Hall coefficient with a negative sign can be seen obviously. The inset shows a good linear relation between ρ_{xy} and magnetic field at different temperatures for sample II.

scattering rate $1/\tau(T)\propto\rho_0(T)$, here f and F represent some unknown functions. For MgB_2, Kohler's law is not obeyed because of the multiband property.[21] Here we tried the scaling based on Kohler's law to the data from the single crystal (SC sample II), the result is shown in the inset of Fig. 2(a). Clearly, the MR data measured at different temperatures do not overlap and Kohler's law is not obeyed in this material. This discrepancy may suggest that in this material there is multiband effect or the basic assumption for Kohler's law $1/\tau(T)\propto\rho_0(T)$ may not be satisfied. Through the following analysis, we will see that the latter may be more plausible.

For a normal metal with Fermi liquid feature, the Hall coefficient is constant versus temperature. However, the Hall coefficient varies with temperature for a multiband material, such as MgB_2,[22] or a sample with non-Fermi liquid behavior such as the cuprate superconductors, e.g., in Ref. 26. In the found iron-based superconductors, R_H varies with temperature in polycrystals.[15,27] People may question that the temperature dependence of R_H observed in polycrystalline samples do not show an intrinsic property, since the electric current flows randomly in different directions on different grains, which gives rise to complexity in interpreting the Hall effect. In the present work, the Hall resistance was carefully measured on $NdFeAsO_{1-x}F_x$ single crystals, and this concern can be removed completely. The inset of Fig. 3 shows the raw data of the transverse resistivity ρ_{xy} at different temperatures, which is in good linear relation against the magnetic field. In Fig. 3, the temperature dependence of the Hall coefficients $R_H=\rho_{xy}/\mu_0 H$ were plotted, which decay continuously with increasing temperature and behave in almost the same way in the two samples. The value of R_H at 400 K is about 20 times smaller than that at 55 K.

A straightforward interpretation to the violation of Kohler's law and the strong temperature dependence of R_H would be the multiband effect, especially if it is true when one assumes an almost balanced contribution of electron and hole pockets. This, however, contradicts the following facts. In a simple two band model, MR can be expressed as follows by omitting the higher-order terms of $B=\mu_0 H$

$$\frac{\Delta\rho}{\rho_0} \simeq \frac{\sigma_1\sigma_2(\mu_1-\mu_2)^2B^2}{(\sigma_1+\sigma_2)^2}, \qquad (1)$$

where $\sigma_i=n_ie^2\tau/m_i$ and $\mu_i=e\tau/m_i$ are the conductivity and the mobility of the ith band, respectively, with n_i and m_i meaning the charge-carrier density and the effective mass of the ith band, respectively. The Hall coefficient can be expressed as

$$R_H \simeq \frac{\sigma_1\mu_1+\sigma_2\mu_2}{(\sigma_1+\sigma_2)^2}. \qquad (2)$$

If assuming the sample that we investigate here has comparable contributions from electron pocket and hole pocket, the magnetoresistance should be quite strong as in MgB_2. This is actually not the case. Band structure calculations[10,13,28] also show that when doping the parent phase ReFeAsO with more than 10% electrons like in our present case, the hole pockets will shrink into small ones and the electron pockets expand a lot. In this case it is very hard to believe that the hole pockets play still an important role in the electric conduction. The two electron pockets surrounding the M-A are highly degenerate, therefore it is natural to assume that they have very similar Fermi velocity and electron mass. The electron scattering may also share high similarities. It is thus very difficult to use Eq. (2) to understand the strong temperature dependence of R_H since the scattering times τ_1 and τ_2 are close to each other, and to the first order assumption no more scattering time is involved in Eq. (2). In order to check whether this is a more reasonable case we did the calculation of $\cot\theta_H=\rho_{xx}/\rho_{xy}\equiv 1/(\omega_c\tau_H)$ which measures only the scattering rate $1/\tau$ for the $NdFeAsO_{0.82}F_{0.18}$ single crystal (here τ_H is the τ determined from Hall angle, ω_c is the circling frequency). Since the temperature dependence of ρ and R_H is very complex and is difficult to be described by the easy expression, we try to find out the relationship between $\cot\theta_H$ and T. Figure 4(a) shows the temperature dependence of $\cot\theta_H$. Then we try to fit the result with the expression of $\alpha+\beta T^n$, and get good fitting results with average exponent $n\sim 2.7$. It is worthwhile to note that the quantity $\cot\theta_H=\rho_{xx}/\rho_{xy}$ measures mainly the scattering time and the information about the charge-carrier density is naturally separated away. This simple power-law-like temperature dependence of $\rho_{xx}/\rho_{xy}\propto 1/\tau_H\propto\alpha+\beta T^n$ is in sharp contrast with the temperature dependence of the in-plane resistivity ρ_{xx}, manifesting strongly that the charge-carrier density may have a strong temperature dependence. We also try to find the temperature dependence of $\cot\theta_H$ for other polycrystalline samples, and it is interesting to see that the exponents from different samples are quite close to each other: $n=2.64\pm0.07$ in $LaFeAsO_{0.9}F_{0.1-x}$ below about 250 K, and $n=2.57\pm0.11$ in $PrFeAsO_{0.89}F_{0.11}$ at temperatures below about 200 K. The inset of Fig. 4(a) shows the Hall angle versus $T^{2.6}$, and the good linear behavior can be observed with different slopes. Therefore the relationship $\cot\theta_H=\alpha+\beta T^n$ ($n=2-3$) may be universal for the iron-based superconductors below some characteristic temperatures.

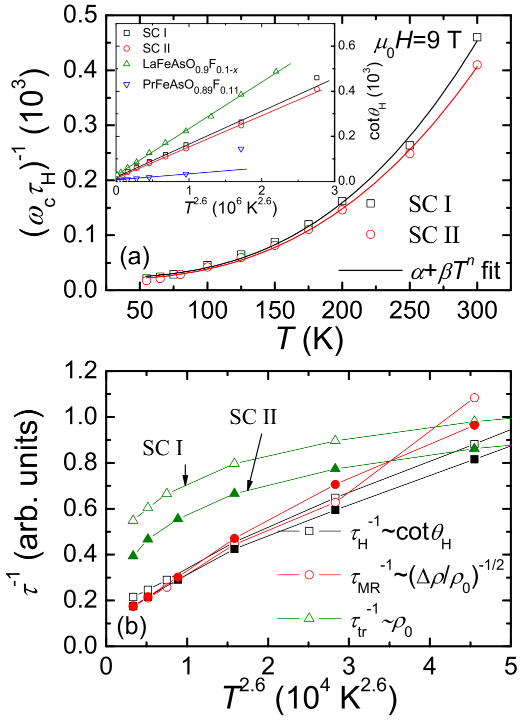

FIG. 4. (Color online) (a) Temperature dependence of $(\omega\tau_H)^{-1}$ of the two samples. The solid lines show the fit by the expression $\alpha+\beta T^n$ with $n=2.82\pm0.11$ for SC sample I and $n=2.62\pm0.08$ for SC sample II. The inset gives the linear behavior between cot θ_H and $T^{2.6}$ for the single crystal and two other polycrystalline samples, and the solid lines show the linear fit to the experimental data (see text). (b) Temperature dependence of τ_H^{-1}, τ_{MR}^{-1}, and τ_{tr}^{-1}.

Bear this idea in mind, we can actually understand the violation of Kohler's law very well. The basic requirement for Kohler's scaling law is $1/\tau(T)\propto\rho_0(T)$, this should not give problems when dealing with a metal with one band and a weak temperature dependent density of states (DOS). However, when the DOS is changing with temperature, Kohler's law is certainly not followed. Assuming that the two electron pockets are the major players of electron conduction in our case, the electrons have slightly different mass m_1 and m_2 on these two bands, but the charge-carrier density n_1 and n_2, and τ_1 and τ_2 are roughly equal to each other, from Eq. (1) we have $\Delta\rho/\rho_0\simeq 1/4(e^2\tau\Delta m/m)^2B^2$ with $\Delta m=m_1-m_2$. Thus one expects that $(\Delta\rho/\rho_0)^{1/2}\propto\tau$. In Fig. 4(b), we accumulate the τ values calculated in different ways. First the $1/\tau_H$ calculated from the Hall angle cot θ_H is proportional to $T^{2.6}$ as mentioned before. This may tell the intrinsic message for the electron scattering in the normal state. Then the $1/\tau_{tr}$ calculated from resistivity exhibits a complicated structure which may reflect a combined result of both the true scattering rate $1/\tau_H$ and the temperature dependent DOS. It is found that the magnetoresistance $(\Delta\rho/\rho_0)^{-1/2}\propto 1/\tau_{\mathrm{MR}}$ scales also with the power law $T^{2.6}$ which suggests $(\Delta\rho/\rho_0)^{1/2}\propto\tau_H$. This consistent analysis reveals that a more reasonable picture to understand the transport data on the single crystals is to assume *a depletion of DOS in lowering temperature*, at least this is part of the reason for the strong temperature dependence of the Hall coefficient R_H.

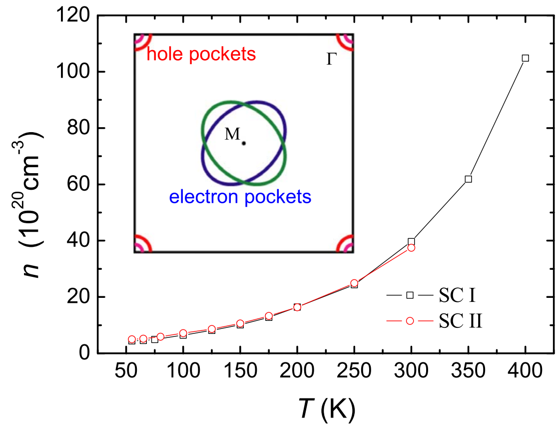

FIG. 5. (Color online) Temperature dependence of charge-carrier density $n=1/R_He$ of the two superconducting samples calculated from R_H based on the single band model. The inset shows the schematic of the Fermi surface (see text) with a heavy electron doping.

The depletion to DOS in lowering temperature is actually a common feature in correlated materials. In cuprate superconductors, this effect is associated with a term "pseudogap" in the normal state, which has been observed by many tools. In the present iron-based superconductors, pseudogap has actually been already inferred from the NMR and photoemission data.[9,29,30] In Fig. 5, based on the single band model (assuming the two electron bands are highly degenerate), we calculate the temperature dependence of the charge-carrier density $n=|1/eR_H|$. It should be mentioned that the temperature dependence of R_H in the iron-based system is much stronger than that in the hole-doped cuprate, where the most accepted picture is the partial gapping of Fermi surface by the pseudogap effect. In the iron-based superconductors, in order to explain the very small magnetoresistance and the strong temperature dependence of the Hall coefficient R_H, the picture concerning partial gapping on the Fermi surface seems necessary.

B. MR and Hall effect of undoped NdFeAsO

The properties of the undoped samples are very different from the superconducting ones, which can be found from the resistive curves shown in Fig. 1. A well accepted picture to understand the resistivity anomaly at about 150 K is the formation of a SDW gap.[16] To analyze the data at temperatures below the SDW transition, we tried the theory given by Andersen and Smith,[31]

$$\rho(T)=\rho_0+AT^2+BT(1+2T/\Delta)\exp(-\Delta/T). \qquad (3)$$

Here, ρ_0 is the residual resistivity, A and B are the fitting parameters; the T^2 term describes the Fermi liquid behavior, while the last term describes the metallic ferromagnet or antiferromagnet state (e.g., Ref. 32) with an energy gap Δ. The curve at temperatures from 40 to 140 K can be well fitted by the expression [see the solid line in Fig. 1(b)], and an SDW energy gap $\Delta\approx 1082$ K is derived.

The SDW state is very interesting and worth further investigation. In Fig. 6 the field dependence of MR and trans-

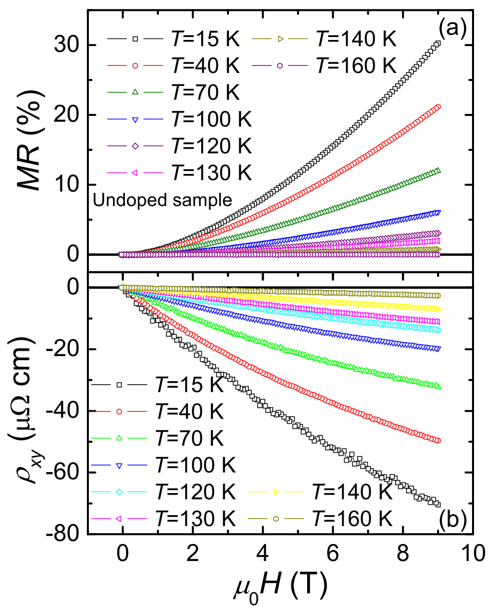

FIG. 6. (Color online) Field dependence of MR (a) and transverse resistivity (b) for the undoped sample at different temperatures. Large MR effect and a little nonlinear Hall effect can be found in the undoped sample.

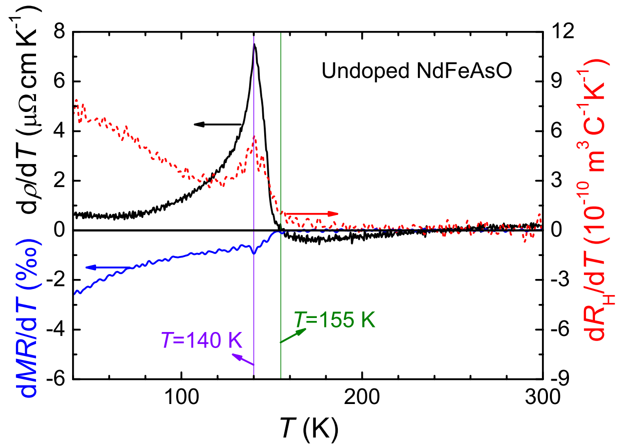

FIG. 8. (Color online) Differential of ρ [from Fig. 1(b)], MR [from Fig. 7(a)], and R_H [from Fig. 7(b)] versus T. Two anomalies could be seen at the temperatures of about 140 and 155 K, which may correspond to the SDW and the structural phase transitions, respectively.

verse resistivity were shown. The MR effect is very strong at low temperatures, and the magnitude is as large as 30% at 9 T and 15 K. In Fig. 6(b), the transverse resistivity shows nonlinear behavior at low temperatures. Both of them show very different behaviors from the superconducting samples. Figure 7 shows the temperature dependence of MR, R_H, and the magnitude of nonlinear Hall effect. Obviously, the curves measured by sweeping temperature at fixed field are consistent with the curves measured by sweeping the field at fixed temperatures, as shown in Fig. 7. All three curves have a clear kink at about 155 K, i.e., at the temperatures below it, both of the values and the changing rates are enhanced gradually, while above that point the values become very small (even vanished) and change slowly with temperature. The large MR and nonlinear Hall effect may be associated with the multiband effect or the complex scattering between the itinerant electrons and the spin moment. The magnetic field will make the long-range spin order (the SDW here) frustrated which leads to more strong scattering to the electrons and thus larger resistivity. In this material, we cannot give a definite judgment about the origin of the strong magnetoresistance, whether it is due to the multiband effect or due to the scattering with magnetic moments. From the temperature dependence of Hall coefficient R_H, one can already see that the transition is accomplished in two steps. A first inflecting point occurs at about 155 K, then it increases in magnitude and another kink appears at a lower temperature, ca. 140 K. The transition at about 155 K may correspond to the structural phase transition, while the lower one at about 140 K may relate to the SDW formation.[16,33] In order to have a close scrutiny on these two transitions, the differential of the ρ-T, MR-T, and R_H-T curves are shown in Fig. 8. Now it becomes very clear that there are indeed two characteristic points at temperatures of around 140 and 155 K, respectively. So the anomalous behaviors of $d\rho/dT-T$, $d\mathrm{MR}/dT-T$, and $dR_H/dT-T$ may start from the structural phase transition, and show a cusplike behavior at the SDW transition temperature. Hopefully a future investigation will give an explanation to this close correspondence.

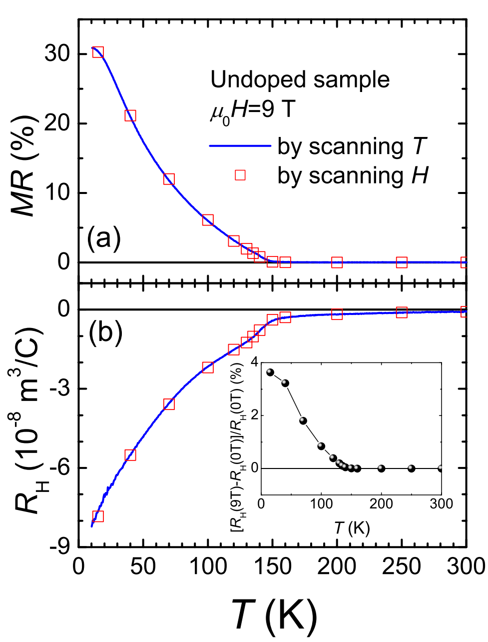

FIG. 7. (Color online) Temperature dependence of MR (a) and Hall coefficient R_H at $\mu_0 H$=9 T. The inset of (b) shows the magnitude of nonlinear Hall effect expressed by $[R_H(9\mathrm{T})-R_H(0\mathrm{T})]/R_H(0\mathrm{T})$.

C. Comparison between the undoped NdFeAsO and the superconducting $NdFeAsO_{0.82}F_{0.18}$

From the results mentioned above, the F doping weakens the SDW and the structural phase transition and finally generates superconductivity beyond a certain doping level.

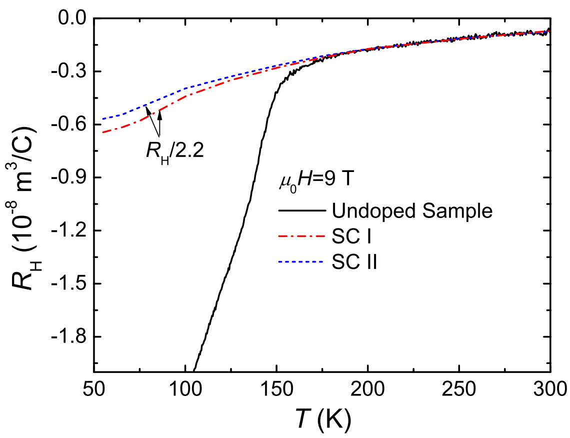

FIG. 9. (Color online) Temperature dependence of R_H of the undoped sample and $R_H/2.2$ of the two superconducting samples. The curves at high temperature above 175 K overlap very well, which suggests that the properties of the samples are very similar to each other in that temperature region.

However, the situation at high temperatures above the two transitions is still unknown. It would be very interesting to have a comparison between properties of these two very different samples. For that the Hall coefficient of the undoped sample and $R_H/2.2$ for two superconducting samples are shown in Fig. 9. Although the absolute value of R_H of the superconducting sample is about two times larger than that of the undoped sample, it is surprising to see that the R_H for two samples can be nicely scaled with a simple multiplication of 2.2 for R_H of the undoped sample from 175 K all the way up to 300 K. This suggests that both the undoped nonsuperconducting samples and the doped superconducting samples may have quite similar Fermi surfaces with different charge-carrier densities in high-temperature region. This gives support to the theoretical calculation that the charge doping may not shift the Fermi energy too much, but rather lift off the very condition for the formation of the SDW.[34] This is a very important conclusion which suggests that the rigid-band model is certainly insufficient to explain the doping effect. On the other hand, one can see that the temperature dependence of resistivity is very different for the two samples in the same temperature region, indicating a very different electron scattering mechanism. A deeper insight to this issue is very interesting and will be carried out in the future investigation.

IV. SUMMARY

In summary, for the first time the MR and Hall effect have been investigated on single crystals of superconducting $NdFeAsO_{0.82}F_{0.18}$ samples and undoped nonsuperconducting NdFeAsO ones. A strong temperature dependence of the Hall coefficient R_H has been found up to 400 K for both samples although a greatly enhanced R_H was observed below about 155 K for the undoped one. It is further found that the MR for the superconducting sample is very weak in the normal state and does not obey Kohler's scaling law. A picture concerning the partial gapping to the density of states at the Fermi surface can consistently explain the dilemmatic phenomenon: strong Hall effect (as well as strong temperature dependence) but very weak magnetoresistance the superconducting sample. For the undoped samples, however, a very strong magnetoresistance was observed in low-temperature region, which could be due to the multiband effect or the electronic scattering with the magnetic moment. The SDW gap about 1082 K for undoped NdFeAsO is derived from the ρ-T curve for the first time. Two step transitions are observed from the transport properties including $\rho(T)$, MR, and $R_H(T)$ which change their behaviors at about 155 and 140 K. These two transitions are in accord very well with the structural phase transition and the SDW formation. The Hall coefficient R_H for both undoped and superconducting samples at temperatures above 175 K show very similar behavior, which suggests the similar Fermi surfaces for both systems and the inapplicability of the rigid-band model in explaining the doping effect.

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