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Inter- to Intra-Layer Resistivity Anisotropy of NdFeAs(O,H) with Various Hydrogen Concentrations

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Abstract

With molecular beam epitaxy and topotactic chemical reaction, we prepared NdFeAs(O,H) epitaxial thin films with various hydrogen concentrations on 5° vicinal-cut MgO substrates. By measuring the resistivities along the longitudinal and transversal directions, the *ab*-plane and the *c*-axis resistivities (ρ_{ab} and ρ_c) were obtained. The resistivity anisotropy $\gamma_{\rho} \equiv \rho_c/\rho_{ab}$ of NdFeAs(O,H) with various hydrogen concentrations was compared with that of NdFeAs(O,F). At the H concentrations which led to superconducting transition temperatures T_c over 40 K, γ_{ρ} recorded ~100-150 at 50 K. On the other hand, a low γ_{ρ} value of 9 was observed with the mostly doped sample. The exponent β of the *ab*-plane resistivity obtained by fitting a power law expression $\rho_{ab}(T) = \rho_0 + AT^{\beta}$ to the data was close to unity down to low temperature in the vicinity where the second antiferromagnetic phase locates, which may be related to the quantum critical point discussed at the over-doped side of the phase diagram.

1. Introduction

Mixed-anion compounds have recently been attracting considerable attention, since they are rich in emergent phenomena, e.g. acid nitride photocatalysts [1] and acid hydride proton conductors [2]. Iron-based mixed-anion compound LnFeAsO (Ln = Nd and Sm) shows superconductivity with a maximum transition temperature T_c of 56 K when oxygen is partially substituted by fluorine $(LnFeAsO_{1-x}F_x)$ or hydrogen $(LnFeAsO_{1-x}H_x)$ [3][4][5]. These substitutions lead to electron doping $(O^{2-} \rightarrow H^{-}/F^{-} + e^{-})$, therefore, the carrier concentration is determined by the amount of F or H. The decisive difference between fluorine and hydrogen is the solubility limit: the latter can be substituted up to $x \sim 0.8$ [6], whereas the former about x = 0.2 at ambient pressure [7]. LnFeAsO crystalizes in a laminated structure with alternating LnO charge reservoir and FeAs conduction layers along the *c*-axis direction. Therefore, the electromagnetic properties of *Ln*FeAsO are anisotropic. From the viewpoint of application, however, a lower electromagnetic anisotropy is favorable. Compared with F-doped LnFeAsO, hydrogen substituted LnFeAsO has a more three-dimensional like Fermi surface, and thus a lower anisotropy can be expected [8]. Additionally, the character of the Fermi surface may depend on the carrier doping level as was shown for another Fe-based superconductor, BaFe₂As₂ [9][10]. Hence, it is interesting to explore how the electromagnetic anisotropy of $LnFeAsO_{1x}H_x$ changes as a function of doping level.

The resistivity anisotropy γ_{ρ} defined as the ratio ρ_c/ρ_{ab} where ρ_c and ρ_{ab} are the out-of-plane and in-plane resistivities, is related to the mass anisotropy $\gamma_m \equiv m_c/m_{ab}$ where m_c and m_{ab} are the out-of-plane and in-plane effective masses, respectively. Polycrystalline *Ln*FeAsO samples with different hydrogen contents have been used for exploring physical quantities [11], but they are not ideal platforms for investigating the electromagnetic anisotropy. Unfortunately though, fabrication of large enough H-doped *Ln*FeAsO single crystals seems to be still difficult, and only a

few studies have been reported measuring the resistivity anisotropy of doped LnFeAsO[12][13][14][15]. Recently, SmFeAsO_{0.9}H_{0.1} single crystals have been grown by combining a Na₃As flux and a high-pressure synthesis method [15]. The γ_{ρ} of the resultant crystals increased from ~3 at 300 K to 8 at 50 K. However, it is not clear how the resistivity anisotropy depends on the hydrogen content, in particular, at high concentration.

Recently, hydrogen doped LnFeAsO epitaxial thin films have been fabricated via a topotactic chemical reaction [16][17][18]. In one of these works, we showed that hydrogen contents can be varied by controlling the processing temperature T_{proc} [18], which enables us to investigate the physical quantities as a function of carrier concentration. Additionally, we have successfully grown off-axis NdFeAsO thin films using vicinal substrates in an earlier work [19], from which the out-of-plane resistivity can be extracted. Combining these two methods, NdFeAs(O,H) thin films having different H concentrations were grown on vicinal-cut MgO substrates in the present study. We measured the transport properties of these films and discussed how the physical quantities and their anisotropy depend on the carrier concentration.

2. Experimental

The growth of parent NdFeAsO thin films with thicknesses of 20-30 nm was carried out by molecular beam epitaxy (MBE) at 800 \Box . Fe, As, NdF₃, Fe₂O₃ and Ga were charged in Knudsen cells. Here, oxygen was supplied via the thermal decomposition of Fe₂O₃, whereas Ga was used to remove F according to NdF₃+Ga \rightarrow Nd+GaF₃. The growth procedure was described in detail in Ref. [20]. In-situ reflection high-energy electron diffraction (RHEED) was utilized for monitoring the film growth. Instead of normal MgO(001) substrates, $5 \times 10 \text{ mm}^2$ sized vicinal-cut substrates with the [001] axis inclined by ~5° with respect to the direction normal to the surface were used for extracting the out-of-plane resistivity [fig. 1(a)]. After the film growth, X-ray diffraction (XRD) analysis was performed: $2\theta/\omega$ scans were used for evaluating the phase purity and the *c*-axis lattice parameter. Because the film grows with an offset angle on a vicinal-cut substrate, we also conducted ω -scans with the angle 2θ fixed to the 003 reflection of NdFeAsO to determine the offset angle α precisely. The angle ω that maximizes the diffraction intensity corresponds to α .

Doping hydrogen into NdFeAsO was conducted similarly to Ref. [18]. As-grown films were cut into smaller samples having a $5 \times 5 \text{ mm}^2$ size. The sample was placed in a Al₂O₃ crucible filled with CaH₂ powder (~0.8 g) as a hydrogen source. The whole arrangement was placed in a quartz tube (outer diameter 12 mm and length 150 mm) and then sealed under vacuum. Importantly, the surface of the film had to be physically in contact with the CaH₂ powder. Then the quartz tube was heated to T_{proc} = 440~500 \Box where it was held for 36 hours. The doping level of H was controlled by adjusting the processing temperature.

The H-doped NdFeAsO films were photolithographically patterned and Ar-ion-beam etched to make two kinds of bridges (50 µm width and 0.5 mm long), as shown in fig. 1(b). They are used for measuring the transport properties in the longitudinal direction (abbreviated as L-bridge) and in the transverse direction (T-bridge), respectively [fig. 1(a)]. A four-probe method was employed with a bias current of 1 µA to measure resistivity using a physical properties measurement system (PPMS). The onset superconducting transition temperature T_c^{onset} was determined as the intersection between the fit to the normal state resistivity and the steepest slope of the transition region. The zero resistivity temperature T_c^0 was defined by a resistivity criterion, $\rho_{cri} = 0.05$ µΩcm, which corresponds to the noise level of our experimental set-up (see Supplementary Material [21]). In the longitudinal direction, the current flows only in the *ab*-plane, so the longitudinal resistivity $\rho_{\rm L}$ is the same as the *ab*-plane resistivity (i.e. $\rho_{ab} = \rho_{\rm L}$), whereas in the transverse direction, the current flows both in the *ab*-plane and along the *c*-axis. Therefore, ρ_c can be calculated from the transverse resistivity ($\rho_{\rm T}$) by the following equation [22]

$$\rho_c = (\rho_{\rm T} - \rho_{\rm L} \cos^2 \alpha) / \sin^2 \alpha \,. \tag{1}$$

The Hall resistivity ρ_{xy} was measured in the field range of up to $\mu_0 H = \pm 9$ T at 50 K. As expected, the slope corresponding to the Hall coefficient $R_{\rm H}$ was always negative, indicating that the dominant carrier is electron. The carrier density *n* was evaluated using the single carrier model formula

$$R_{\rm H} = -\frac{1}{ne}.$$
 (2)

Here, *e* is the elementary charge. Note that for the Hall effect measurements, the bias current was supplied along the longitudinal direction, whereas the Hall voltage was measured along the transverse direction, which is the tilted direction. This may lead to a deviation between the actual carrier density and the measured one [23]. The upper critical field H_{c2} was determined by measuring the field dependence of resistivity and using a criterion of 90% of the normal state resistivity.

3. Results and discussion

3.1 Structural characterization

The specifications of the samples studied here are listed in table 1. For convenience, these samples are named in the order of their carrier densities. We used five as-grown films. Samples B and E were prepared from the same as-grown film and all other samples were prepared from different as-grown films. As stated above, the doping level of H can be controlled by adjusting the processing temperature T_{proc} . However, this applies only to the samples derived from the same as-grown film. That is why the correlation between *n* and T_{proc} seems to be weak (see table 1).

XRD patterns of samples B and E, both originating from the same as-grown film, are compared in fig. 2. For comparison, the pattern of the as-grown film is also shown. Although small peaks arising from impurities were observed for sample B, the XRD patterns of all samples including those not shown here dominantly consist of the 00*l* peaks of the NdFeAs(O,H) phase and the substrate peaks, indicating that they are *c*-axis oriented. The intensity of the 002 peaks of the doped samples in fig. 2 were small and almost indistinguishable from the background. Our study using an ordinary (not a vicinal cut) substrate shows that the intensity of the 002 peak relative to the other peaks decreases with H doping (see Supplementary Material [21]). Hence, the weaker intensity is, at least partially, due to the slight change in the crystal coordinates and the difference in the atomic scattering factors of oxygen and hydrogen. However, a slight degradation of the crystallinity cannot be ruled out completely. By comparing the doped samples with the as-grown one, clear shifts of the 00*l* peaks to higher angles are recognized, which reflects the reduction in the *c*-axis length. The evaluated *c*-axis lengths for all samples are summarized in table 1. The carrier density and the *c*-axis length are well correlated, which is consistent with our previous results on NdFeAs(O,H) grown on ordinary MgO(001) substrates [18] and found in bulk H-doped or F-doped *Ln*FeAsO samples [24][25].

3.2 Resistivity measurements at zero magnetic field

The temperature dependencies of the resistivity in the transversal and longitudinal directions for all samples are summarized in fig. 3(a). As can be seen, the transversal resistivity is always larger than

the longitudinal one because ρ_T contains the ρ_c component. For all samples, each pair of transversal and longitudinal bridges fabricated from the same film possess the same T_c^{onset} and T_c^0 , proving that the films are chemically homogeneous.

The carrier density dependencies of both T_c^{onset} and T_c^0 are plotted in fig. 3(b). Both T_c^{onset} and T_c^0 decreased with increasing carrier concentration. As the behavior of T_c^{onset} and T_c^0 on *n* are similar, T_c^{onset} is regarded as T_c from here on for simplicity.

The temperature dependencies of the in-plane resistivity $\rho_{ab}(T)$ and the out-of-plane resistivity $\rho_c(T)$ of all samples are plotted in figs. 4(a) and (b), respectively. All $\rho_{ab}(T)$ curves exhibited a metallic behavior irrespective of *n*. However, the shape of the $\rho_{ab}(T)$ curve for samples E and F is obviously different from that of samples A, B and C. The $\rho_{ab}(T)$ curves of the former samples have a positive curvature, while those of the latter samples are slightly negative. On the other hand, sample E shows an almost linear temperature dependence from T_c to 120 K (see Supplementary Meterial [21]). This will be discussed later. The behavior of $\rho_c(T)$ shown in fig. 4(b) is converse to that of $\rho_{ab}(T)$. ρ_c depends almost linearly to temperature at larger *n*, while the $\rho_c(T)$ curves of the less doped samples have a negative curvature. Moreover, the temperature dependence of ρ_c for samples A-C showed a peak at around 250-275 K, which may correspond to a crossover from incoherent conduction at high temperatures to coherent conduction at low temperatures as was observed, for instance, also in the BaFe₂As₂ system [26]. Note that the correlation length *L* evaluated by the Williamson-Hall analysis is close to the film thickness (20~30 nm) and independent of carrier concentration [27](see Supplementary Material [21]). Thus, the variation of ρ_c is not due to the change in the microstructure but the H content.

Figure 4(c) shows the *n* dependence of ρ_{ab} at 300 and 50 K. ρ_{ab} at both 300 and 50 K decrease with increasing *n* up to 4×10^{21} /cm³. An abrupt rise in ρ_{ab} at around $n = 4 \times 10^{21}$ /cm³ is observed at both 300 and 50 K. This carrier concentration coincides with that where a quantum critical point (QCP) may be present as will be discussed later. Unlike ρ_{ab} , ρ_c at 300 and 50 K decrease monotonically with increasing *n*, as shown in fig. 4(d).

The resistivity anisotropy γ_{ρ} is shown in fig. 5(a) as a function of temperature. For samples A-E, γ_{ρ} increases monotonically with decreasing *T*. This behavior is the same as that of optimally F-doped NdFeAsO thin films [19]. The maximum γ_{ρ} of the samples with a T_c above 40 K reaches about 100 to 150 just above T_c . The temperature dependence of γ_{ρ} becomes weaker and the value of γ_{ρ} decreases as *n* increases. Finally, γ_{ρ} of the mostly doped sample has a different temperature dependence. It decreases with decreasing temperature and records a minimum of about 8 around 100 K before increasing at lower temperatures as shown in fig. 5(b). Although the variation in the temperature dependence of the present sample F is small, the minimum of γ_{ρ} might be an indication of a phase transition at around 100 K. In fact a structural transition was observed for SmFeAs(O,D) and LaFeAs(O,H) at about 100 K in the over-doped region, where the *c*-axis length showed a minimum [6][28]. As a shorter *c*-axis length means an increased connectivity between the FeAs conducting layers, this may explain the minimum of the resistivity anisotropy.

Figure 5(c) shows the carrier density dependence of γ_{ρ} at 50 K for H-doped NdFeAsO. The films with higher T_c tend to have a larger anisotropy. The peak of γ_{ρ} is located at around $n = 2.2 \times 10^{21}$ /cm³, which corresponds to the high- T_c region [see fig. 3(b)]. It is notable that the optimally F-doped sample has a larger anisotropy than the H-doped samples [19]. This might be explained by the more two-dimensional character of the Fermi surface of F-doped samples [8]. Alternatively, the high reactivity of fluorine may have introduced some damage to the crystal during the fluorination

that disturbed the interplane conductivity.

3.3 In-field resistivity measurements of samples C and E

The upper critical fields H_{c2} of sample E determined from the in-field ρ_{ab} measurements for both $H \parallel c$ and $\parallel ab$ are summarized in fig. 6(a). The respective slopes of the temperature dependence are -4.8 T/K and -14.8 T/K near T_c for $H \parallel c$ and $\parallel ab$, which gives an anisotropy of 3.1. For single-band superconductors, the anisotropy of the upper critical field $\gamma_{H_{c2}} (= H_{c2\perp}/H_{c2\parallel})$, $H_{c2\perp}$ and $H_{c2\parallel}$ are the upper critical fields for $H \parallel ab$ and $\parallel c$) equals to $\sqrt{\gamma_{\rho}} (= \sqrt{\gamma_m})$. This relation does not necessary hold for multi-band superconductors. Nevertheless, the value of $\gamma_{H_{c2}}$ is relatively close to the square root of the resistivity anisotropy [γ_{ρ} (50 K) ~ 15] of sample E.

We also measured the resistivity under magnetic fields in the flux-flow regime of sample E, and found that the angle dependence of resistivity under various magnetic fields collapses to a single curve when plotted as a function of an effective magnetic field $\mu_0 H_{\text{eff}} = \mu_0 H/f_{\text{T}}(\theta)$, where $f_{\text{T}}(\theta)$ is the Tinkham formula given by [29]

$$f_{\rm T}(\theta) = \frac{1}{2\gamma_{H_{\rm c2}}{}^2 \sin^2\theta} \left[\sqrt{\cos^2\theta + 4\gamma_{H_{\rm c2}}{}^2 \sin^2\theta} - |\cos\theta| \right]. \tag{3}$$

Here, θ is the angle between the magnetic field and *c*-axis. Figure 7 shows the data taken at 26.5 K. The anisotropy parameter $\gamma_{H_{c2}}$ was determined as 3.2±0.1 from the scaling of fig. 7(b), consistent with the value obtained from the temperature slope of H_{c2} . It is also noticeable that the dip in the resistance curve is shifted from 270° to a higher angle at low magnetic fields due to the tilted growth of the film and the corresponding non-parallelity of B and H (see fig. 7(c)) [30]. The shift of the resistance dip explains the slight deviation of the data near zero effective field from the scaling curve in fig. 7(b). Resistivity scaling at slightly higher temperatures (27.0 and 27.5 K) with the same value of $\gamma_{H_{c2}}$ were of similar quality. On the assumption that the relaxation time of carriers is isotropic, the resistivity anisotropy is equal to the mass anisotropy. In our previous study, the mass anisotropy was different in the normal and superconducting states for under- and optimally doped NdFeAsO [19]. Hence, the rough match of the mass anisotropy in the normal state with the one in the superconducting state may be one of the distinct features of the over-doped side of the phase diagram. Fig. 6(b) compares H_{c2} of sample E with that of a less doped one (sample C). For $H \parallel ab$, H_{c2} of both samples are almost identical. On the other hand, for $H \parallel c$, sample E shows a higher H_{c2} at the same reduced temperature T/T_c . This is probably due to an increased coupling between FeAs layers with increasing doping.

3.4 Exponent of power law behavior $\rho_{ab}(T) = \rho_0 + AT^{\beta}$

In order to quantify the behavior of the temperature dependence of ρ_{ab} , we examined the exponent β by assuming $\rho_{ab}(T) = \rho_0 + AT^{\beta}$ in the temperature range from just above T_c to 120 K, where ρ_0 is the residual resistivity at zero kelvin and A is a constant. The obtained β is plotted against the carrier density in fig. 8(a), which shows a dome-like shape. A similar behavior was also found for polycrystalline *Ln*FeAs(O,H) [11].

To investigate the exponent β in more detail, we employed the formula $\beta(T) = \frac{d\ln(\rho_{ab} - \rho_0)}{d\ln T}$ to

extract the temperature dependence of β over a wide temperature range [fig. 8(b)], where the ρ_0 value obtained by the fitting described above was used. For all samples, β is close to 2 at the temperature very close to T_c , indicative of a Fermi liquid state. A similar behavior was also found in

FeSe_{0.89}S_{0.11} under various pressures [31]. At higher temperatures, β is smaller than 2. For example, β is almost constant around 1.5 for a wide temperature range from 200 K to close to T_c as shown in fig. 8(b) for sample B, consistent to the result of the fitting mentioned above.

At $n \approx 4.2 \times 10^{21}$ /cm³, on the other hand, β was close to 1 over the temperature range from 150 K to T_c , and the general trend of the resistivity curve changed from positive ($\beta > 1$) to negative ($\beta < 1$) curvature with further increasing *n*. Intriguingly, a rather abrupt change in the ρ_{ab} value was observed at this carrier density as pointed out above (fig. 4(c)). The *T*-linear behavior of resistivity has often been associated with a quantum critical point at T = 0. However, the *T*-linear behavior usually extends to a fan-like region around the quantum critical point on the phase diagram [32], although here it is observed only in a strip-like region. The electronic phase diagram of H-doped LaFeAsO reported by Hiraishi *et al.* shows that the second antiferromagnetic phase (AFM2) and superconductivity may coexist in the over-doped region [28]. The Néel temperature decreases with the application of a high pressure, and the presence of a pressure-induced quantum critical point (QCP) was suggested from NMR measurements [33]. The anomalous behaviors we have observed at $n \approx 4.2 \times 10^{21}$ /cm³ might be a glimmering of this QCP at ambient pressure, although whether they are indeed related or not is an open question for future studies. Note though that the actual carrier density might be slightly different as mentioned in the experimental section.

For larger values of *n*, β is less than 1 in the whole temperature range. For a SmFeAsO_{0.9}H_{0.1} single crystal, an exponent $\beta = 0.47$ was reported and associated to the spin-frozen state that locates between AFM1 and the superconducting phases [15]. The electronic phase diagram of *Ln*FeAsO shows that AFM1 and AFM2 exist close to the superconducting phase in the under-doped and the over-doped regimes, respectively [6][28]. It might be possible to explain the small β value in the over-doped region similarly.

4. Conclusion

NdFeAs(O,H) epitaxial thin films with different doping concentrations were grown on vicinal-cut MgO substrates by MBE. With the combination of F-doped samples in the lower carrier density regime, a large part of the phase diagram can be mapped. The temperature dependence of both ρ_{ab} and ρ_c exhibits metallic behavior irrespective of the doping level for all samples studied in this work. The extracted resistivity anisotropy γ_{ρ} showed a strong doping dependence. At the H concentrations which led to superconducting transition temperatures (T_c) over 40 K, γ_{ρ} recorded ~100-150 at 50 K. With increasing the carrier concentration, γ_{ρ} eventually reached a remarkably small value around 8. We extracted the *T*-dependent exponent β of $\rho_{ab}(T)$ and discussed the presence of a possible QCP at the over-doped side of the phase diagram.

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- [21] See Supplemental Material at [], we describe (i) Determination of the onset superconducting transition temperature T_c^{onset} and zero resistance temperature T_c^0 , (ii) 20/ ω scans of NdFeAs(O,H) films grown on CaF2 substrates, (iii) Resistivity curves $\rho_{ab}(T)$, and (iv) Williamson-Hall plot for analyzing the correlation length.
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Figure 1. (a) Schematic illustration of a vicinal-cut substrate. The [001] axis is tilted from the direction normal to the substrate surface by θ_{vic} . (b) An example of a top-view optical micrograph of the micro-bridges. A bridge running along the transverse direction is labeled as a T-bridge, whereas a bridge parpendicular to the transverse direction is labeled as an L-bridge.



Figure 2. The $2\theta/\omega$ scans of samples B (blue) and E (red), which were processed at 460°C and 470°C, respectively. For comparison, the $2\theta/\omega$ scan of a pristine film (black) with no doping is also shown.

Table 1. Sample name, carrier density n , processing temperature T_{proc} of topotactic reaction, the
<i>c</i> -axis lattice parameter, the growth angle (offset angle of the sample) α , and the onset T_c (T_c^{onset}) of
the samples of this study. The samples are named in the order of the carrier density for convenience.

$n (10^{21} / \text{cm}^3)$	$T_{\text{proc}}(\Box)$	<i>c</i> -axis (Å)	α (°)	$T_{\rm c}^{\rm onset}({\rm K})$
1.51	440	8.482 ± 0.009	5.75	50.5
2.28	460	8.418±0.016	5.81	42.1
2.75	450	8.418±0.003	4.95	43.1
3.99	500	8.424±0.012	6.21	37.4
4.23	470	8.386±0.005	5.92	27.8
5.03	470	8.361±0.004	5.93	24.3
	n (10 ²¹ /cm ³) 1.51 2.28 2.75 3.99 4.23 5.03	$n (10^{21} / \text{cm}^3)$ $T_{\text{proc}} (\Box)$ 1.514402.284602.754503.995004.234705.03470	$n (10^{21} / \text{cm}^3)$ $T_{\text{proc}} (\Box)$ c -axis (Å)1.51440 8.482 ± 0.009 2.28460 8.418 ± 0.016 2.75450 8.418 ± 0.003 3.99500 8.424 ± 0.012 4.23470 8.386 ± 0.005 5.03470 8.361 ± 0.004	$n (10^{21} / \text{cm}^3)$ $T_{\text{proc}} (\Box)$ c -axis (Å) α (°)1.51440 8.482 ± 0.009 5.75 2.28460 8.418 ± 0.016 5.81 2.75450 8.418 ± 0.003 4.95 3.99500 8.424 ± 0.012 6.21 4.23470 8.386 ± 0.005 5.92 5.03470 8.361 ± 0.004 5.93



Figure 3. (a) The temperature dependencies of $\rho_{\rm L}$ (copper) and $\rho_{\rm T}$ (green) for samples A-F. (b) $T_{\rm c}^{\rm onset}$ and $T_{\rm c}^{\rm 0}$ as a function of the carrier density for NdFeAs(O,H).



Figure 4. (a-b) The temperature dependence of ρ_{ab} (a) and ρ_c (b) of samples A (green), B (fuchsia), C (red), D (blue), E (copper) and F (lime). (c-d) The carrier density dependencies at 300 and 50 K for (c) ρ_{ab} and (d) ρ_c , respectively. For sample A, the resistivities (ρ_{ab} and ρ_c) at 50 K were determined by extrapolating the result of linear regression in the temperature range of 55 K < T < 65 K to T = 50 K for its high T_c^{onset} (= 50 K). The solid line is a guide to the eye.



Figure 5. (a) The temperature dependence of the resistivity anisotropy $\gamma_{\rho}(T) = \rho_c/\rho_{ab}$ of samples A-F. (b) An enlarged view of $\gamma_{\rho}(T)$ of sample F. (c) The carrier density dependence of $\gamma_{\rho}(T)$ of NdFeAs(O,H).



Figure 6. (a) Temperature dependence of H_{c2} of sample E for $H \parallel ab$ and $\parallel c$. The solid lines show the results of linear fitting to the data up to 8 T. (b) Comparison between samples C and E.



Figure 7. (a) Angular dependence of the resistance at 26.5 K. The magnetic fields were increased by 0.5 T up to 5.0 T and by 1.0 T further on. A shift of the resistance dip from $H \parallel ab$ towards the substrate surface direction (*s*) at low magnetic fields due to non-parallelity of *B* and *H* is clearly observed. (b) 2D (Tinkham) scaling of the data shown in (a). An anisotropy value of $\gamma_{H_{c2}} = 3.2 \pm 0.1$ yielded the best result. (c) Schematic illustrations of how the direction of *B* changes with the strength of the applied magnetic field. When a weak magnetic field is applied parallel to the substrate, *B* is almost parallel to the crystallographic *ab*-plane due to the trapping of *B* along the Nd(O,H) blocking layers. On the other hand, as a strong magnetic field is applied, the direction of *B* is parallel to that of *H*.



Figure 8. (a) Relationship between the exponent β and *n*. The exponent β was obtained by fitting the power-law form $\rho(T) = \rho_0 + AT^{\beta}$ to the resistivity data in the temperature range of $T_c < T \le 120$ K. The dashed line indicates $\beta = 1$. (b) The carrier density–temperature phase diagram. The colour map represents the exponent β .