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The Hall effect and thermoelectric power correlated with the giant magnetoresistance in modified FeRh compounds

Y Kobayashi[1], **K Muta and K Asai**

Department of Applied Physics and Chemistry, The University of Electro-Communications, Chofugaoka 1-5-1, Chofu-shi, Tokyo 182-8585, Japan

E-mail: koba@pc.uec.ac.jp (Y Kobayashi)

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Abstract

Simultaneous measurements of the magnetoresistance, magnetization, Hall effect, thermoelectric power and specific heat of a modified FeRh compound where 3.5 at.% Ni is doped at the Fe sites are investigated in order to elucidate the origin of the giant-magnetoresistance phenomena through the metamagnetic transition from the antiferromagnetic (AF) to the ferromagnetic (F) state. The Hall coefficient changes its sign through the metamagnetic transitions. The sign of the thermoelectric power also changes from positive to negative through the transition from the AF to the F state. The electronic specific heat coefficient increases remarkably through the metamagnetic transition from the AF to the F state. These results imply that the giant-magnetoresistance effect in FeRh compounds results from the reconstruction of the Fermi surface across the metamagnetic transition. At low temperatures, two-step metamagnetic transitions were found, suggesting an uncompensated AF or ferrimagnetic state in the intermediate-field region around 2–3 T, only in the process of increasing the field.

1. Introduction

There has been great interest in the magnetic and electrical transport properties of ordered FeRh compounds for more than fifty years. The ordered FeRh system has a CsCl-type structure. The system has been reported to show a first-order phase transition from an antiferromagnetic (AF) ground state at low temperature to a ferromagnetic (F) state around the critical temperature $T_0 = 330$ K [1–3]. In the AF state below T_0, a magnetic field induces a first-order metamagnetic transition to the F state [4]. The critical temperature and the field of the transition change on substitution at Fe or Rh sites [5, 6]. The temperature dependence of the resistivity shows a large jump around T_0 [7]. Through the metamagnetic transition, a giant-magnetoresistance (GMR) effect is observed [5].

[1] Author to whom any correspondence should be addressed.

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Two mechanisms are usually considered as origins of the GMR effect associated with the transition between AF and F states. One possible mechanism is a spin-dependent scattering of conduction electrons, which leads to a reduction of the relaxation time in the AF state. The mechanism is considered to produce GMR commonly in magnetic multilayer systems [8]. The FeRh system can be regarded as a naturally occurring magnetic multilayer which consists of alternating monolayers of magnetic iron and nonmagnetic rhodium atoms stacked in the [111] direction of the CsCl unit cell, and shows the GMR effect through the transition from the AF to the F state [9]. On the basis of these facts, some researchers reported that the origin of the GMR in the FeRh compound is the spin-dependent scattering of the conduction electrons [10, 11]. However, it may not be reasonable to assume the scattering to be due to the disorder at interlayer boundaries and the spin-split density of states for each Fe monolayer. Another possible mechanism of GMR is the variations of the Fermi surface through the metamagnetic transition, e.g., due to the formation of the superzone gap in the AF state.

In order to make clear the origin of the GMR in FeRh compounds, measurements of the transport properties such as the Hall effect and the thermoelectric power are desirable. The Hall resistivity in magnetic materials is described as a sum of two terms: the normal Hall resistivity and the extraordinary one. The normal Hall effect contains information on the Fermi surface such as the carrier concentration and/or the $\boldsymbol{k}$-dependence of the conduction electron scattering [12]. The extraordinary Hall effect provides information on the left–right asymmetry of the scattering [12]. The thermoelectric power reflects the energy derivative of the density of states and that of the relaxation time of the conduction electrons [13].

In this paper, we report on simultaneous measurements of the resistivity, Hall effect, thermoelectric power and magnetization for a modified FeRh compound where 3.5 at.% Ni is doped at Fe sites. In an unmodified FeRh compound, the critical field (H_c) is too large below 300 K for one to measure the field dependence of the magnetic and the transport properties through the metamagnetic transition. In order to enable such measurements, we prepared a $(Fe_{1-x}Ni_x)_{49}Rh_{51}$ ($x = 0.035$) compound [5]. To obtain the change of the density of states at the Fermi level through the metamagnetic transition, the specific heat measurement was also performed up to $\mu_0 H = 7$ T.

2. Experimental procedure

The $(Fe_{1-x}Ni_x)_{49}Rh_{51}$ ($x = 0.035$) compound (we denote it as Fe(3.5% Ni)Rh in the following sections) was prepared by melting the components in a tri-arc furnace under an argon atmosphere. After melting, the ingots were annealed in high vacuum ($<5 \times 10^{-6}$ Torr) for 50 hours at 1000 °C and cooled to room temperature at a rate of 1 K min^{-1} [5]. The ordered CsCl-type structure was confirmed by x-ray diffraction measurement. The compositions of Fe, Ni and Rh were checked by an electron-probe microanalyser (JEOL, JAX-8800R). The sample was shaped into thin plates about $5 \times 3 \times 0.4$ mm^3 in size.

The Hall and the magnetoresistive voltages were measured by a conventional dc four-probe method using a computer-controlled current source and an HP 34420A nanovoltmeter. The thermoelectric power was measured by a differential method using AuFe–normal-silver thermocouples. The magnetic field was applied using a superconducting magnet, with a field of up to 9 T. Magnetization measurements were performed up to 7 T using a Quantum Designs SQUID magnetometer. The specific heat was measured by a relaxation method using a Quantum Designs PPMS. The magnetic field was applied perpendicularly to the sample plane for the transport and magnetic measurements, and was applied in the plane for the specific heat measurement.

3. Experimental results

3.1. Magnetoresistance and Hall effect

Figure 1 shows the field dependence of the Hall resistivity (ρ_H), electrical resistivity (ρ) and magnetization (M) at 200 K. At this temperature, the sample is ferromagnetic at any field and shows neither the metamagnetic transition nor the GMR effect. $\rho_H(H)$ mimics $M(H)$ for ordinary ferromagnetic metals. For ordinary ferromagnetic metals, $\rho_H(H)$ is described empirically as a sum of two terms: the normal Hall resistivity R_0H and the extraordinary one R_sM where R_0 and R_s are the normal and the extraordinary Hall coefficients, respectively [12]. In the FeRh system, $\rho_H(H)$ in the F state can be decomposed into the normal and the extraordinary Hall effects (ρ_H^M) resulting from the magnetic scattering as follows. The normal Hall coefficient R_0 in the F state is estimated as $R_0 = (0.26 \pm 0.03) \times 10^{-9}$ m^3 C^{-1} from the slope of $\rho_H(H)$ above 1.5 T where $M(H)$ almost saturates[2]. In contrast, at 5 K where the sample is in the AF state at zero field, $M(H)$, $\rho(H)$ and $\rho_H(H)$ show a complex field dependence as shown in figure 2. The field dependence of M for increasing field shows a plateau around 2–3 T and abruptly increases at around 3–5 T. This two-step metamagnetic transition suggests the existence of an uncompensated AF or ferrimagnetic state in the intermediate-field region around 2–3 T. Above about 6 T, $M(H)$ almost saturates at about 4 μ_B showing that the sample is in the F state. In the increasing-field process, $\rho(H)$ slightly increases at the first metamagnetic transition and shows a huge drop at the second metamagnetic transition. The slope of $\rho_H(H)$ is negative up to about 2 T, and changes its sign to positive in the region where $M(H)$ shows a plateau. In the region of the second metamagnetic transition, $\rho_H(H)$ shows a complex field dependence. Above 6 T where both $M(H)$ and $\rho(H)$ saturate, the slope of $\rho_H(H)$ is constant with positive sign, and the magnitude of the slope suggests R_0 for the F state at 5 K. These facts show that the Hall coefficient changes remarkably in association with the metamagnetic transitions, suggesting a change of the Fermi surface. In the AF and intermediate states below 3 T, $\rho_H^M(H)$ must change with increasing field since $M(H)$ changes. Therefore, it is impossible to decompose ρ_H into the normal and the extraordinary components. However, the remarkable changes of both the magnitude and the sign of the Hall coefficients in the intermediate-field region where $M(H)$ changes only a little suggest that the Fermi surface differs even between the AF and intermediate states. Accordingly, the results of the present experiment suggest that the origin of the GMR effect in FeRh systems is a reconstruction of the Fermi surface resulting from the metamagnetic transition.

The value of R_0 at 5 K is almost the same as that at 200 K, implying that R_0 in the F state is almost temperature independent. On the other hand, the magnitude of the extraordinary Hall component in the F state at 5 K, which is obtained from the intercept of $\rho_H(H)$ above 6 T with the vertical axis (see figure 2(a)), is much smaller than that at 200 K. In order to investigate the characteristics of the magnetic scattering in the F state, we analyse the temperature dependence of the extraordinary Hall component. Figure 3 shows the temperature dependence of ρ_H, ρ and M. In both $\rho(T)$ at $\mu_0H = 0$ T and $M(T)$ at $\mu_0H = 0.1$ T, large hysteresis suggesting a first-order transition between AF and F states is clearly seen. On the other hand, in high magnetic field, the transition vanishes and the sample is ferromagnetic in the temperature range investigated. In figure 3(a), the normal component $R_0H(T)$ and the extraordinary component $\rho_H^M(T)$ at 9 T are also shown in addition to the measured $\rho_H(T)$. $\rho_H^M(T)$ was obtained by subtracting the normal part R_0H estimated from the slope of $\rho_H(H)$ in higher fields at each temperature. The $\rho_H^M(T)$ thus obtained increases remarkably with increasing temperature, as

[2] R_0 derived by the present procedure is subject to error due to the increase of $M(H)$ in the saturation region above 1.5 T. The error is estimated to be at most 0.03×10^{-9} m^3 C^{-1}.

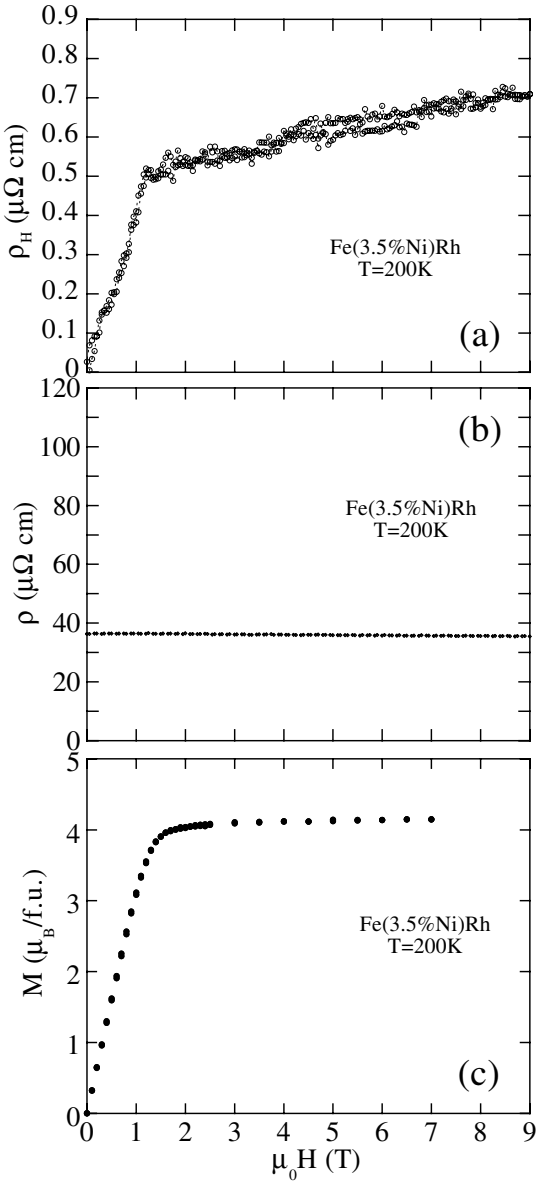

Figure 1. The field dependence of (a) the Hall resistivity (ρ_{H}), (b) the electrical resistivity (ρ) and (c) the magnetization (M) of Fe(3.5% Ni)Rh at 200 K.

is usually observed in ordinary ferromagnetic metals. The origin of the left–right asymmetric scattering responsible for the extraordinary Hall effect can be classified into two mechanisms: the skew scattering proportional to ρ and the side-jump scattering proportional to ρ^2 [12]. Accordingly, the extraordinary Hall coefficient R_s for the ferromagnetic state can be described as

$$R_s = a\rho + b\rho^2 \tag{1}$$

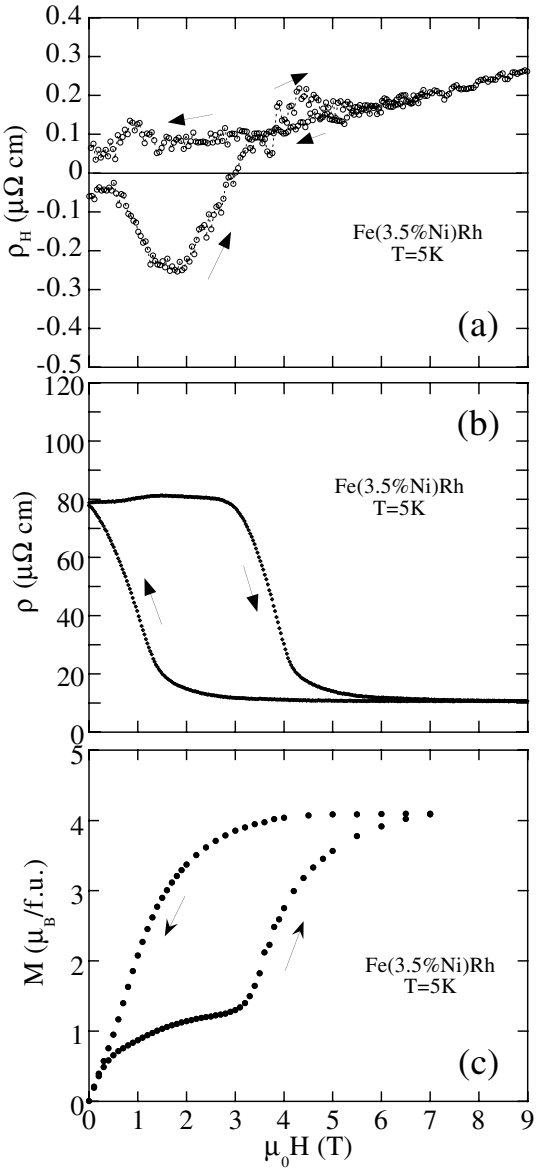

Figure 2. The field dependence of (a) ρ_{H}, (b) ρ and (c) M at 5 K.

or

$$R_s/\rho = a + b\rho. \tag{2}$$

If equation (2) holds, the R_s/ρ versus ρ plot in the ferromagnetic state should be on a straight line, and the contribution of the skew and the side-jump scattering mechanisms can be separated from the plot. To analyse the extraordinary Hall effect in the ferromagnetic state, we plot R_s/ρ versus ρ as shown in figure 4. The R_s/ρ data for the ferromagnetic state are linear against ρ, though the scatter of the data points is fairly large due to the experimental error. Thus we conclude that both the skew and the side-jump scattering contribute to the extraordinary

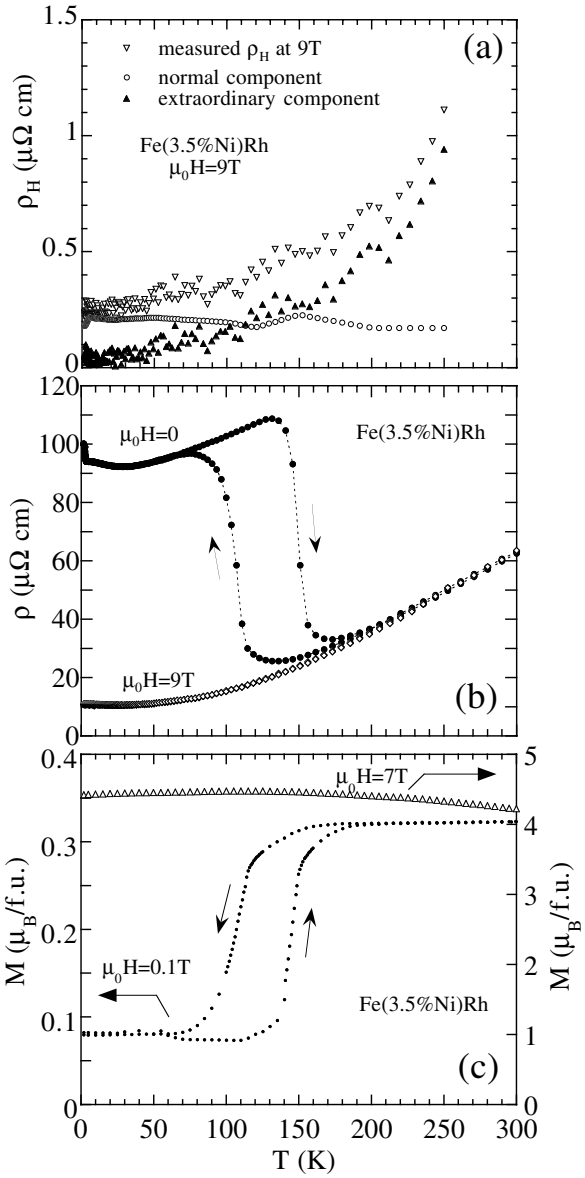

Figure 3. The temperature dependence of (a) ρ_{H}, (b) ρ and (c) M. In (a), the normal Hall component $R_0H(T)$ determined from the high-field slope of the measured $\rho_{\mathrm{H}}(H)$ at each temperature and the remaining extraordinary component $\rho_{\mathrm{H}}^{M}(T)$ $(=\rho_{\mathrm{H}}(T) - R_0H(T))$ at 9 T are also shown.

Hall effect in the ferromagnetic state in FeRh systems, as observed in ordinary ferromagnetic materials [12]. The parameters can be deduced from the fit of the data to equation (2) as $a = (3.2 \pm 0.5) \times 10^{-3}$ T^{-1} and $b = (1 \pm 0.5) \times 10^{-4}$ $\mu\Omega$ cm^{-1} T^{-1}, respectively, which are of the same order of magnitude as those observed for ordinary ferromagnetic metals such as Fe and Co [14]. This result suggests itinerant-electron metamagnetism in FeRh systems including the polarized 3d band and the 4d one, because the large side-jump scattering of the conduction electrons takes place only in itinerant-electron ferromagnetism [12].

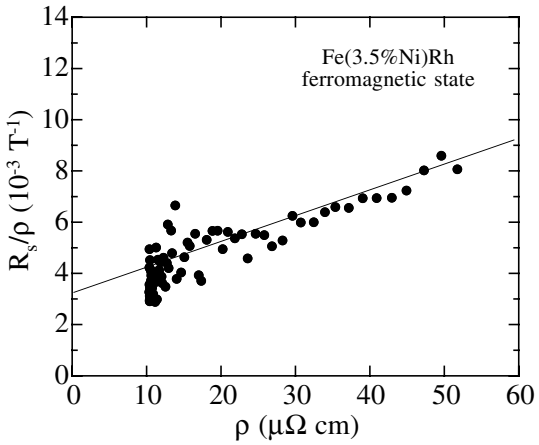

Figure 4. A R_s/ρ versus ρ plot for the ferromagnetic state, with the temperature as an implicit parameter.

3.2. Thermoelectric power

Figure 5 shows the temperature dependence of the thermoelectric power (S) for $\mu_0 H = 0$ and 9 T. At zero field, a large hysteresis was observed in $S(T)$, in accord with the first-order transition. In the F state at higher temperatures, the sign of S is negative. Around the critical temperatures, $T_0 = 100$ K for decreasing temperature and 145 K for increasing temperature, abrupt sign changes of S occur. At $\mu_0 H = 9$ T where the sample is ferromagnetic over the whole temperature range investigated, the sign of S is negative and S smoothly varies as a function of temperature. For higher temperatures, the values of $S(T)$ are almost identical to

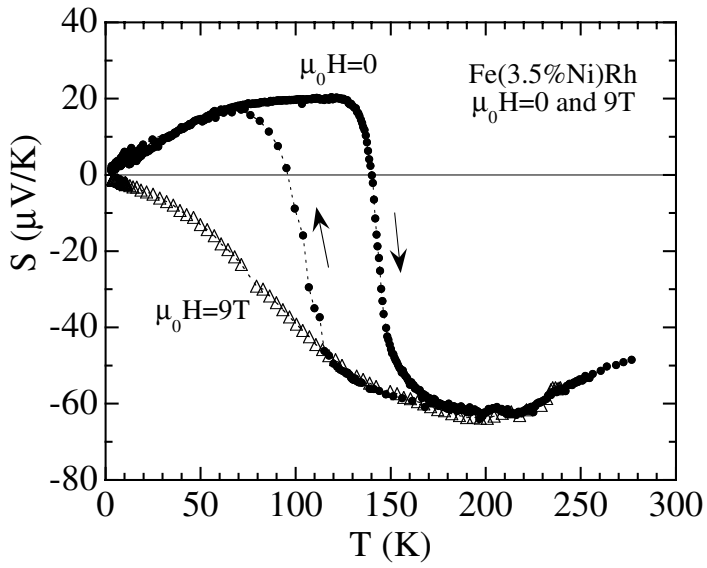

Figure 5. The temperature dependence of the thermoelectric power (S) measured at $\mu_0 H = 0$ and 9 T.

that in the F state at zero field . The sign changes of S between $\mu_0 H = 0$ and 9 T below T_0 were consistent with the field dependence of S at $T = 8$, 55 and 98 K, as shown in figure 6. According to the Mott equation for thermoelectric power,

$$S = -\frac{\pi^2 k_B^2}{3|e|} T \left[\frac{\partial \ln N(E)}{\partial E} + \frac{\partial \ln \tau(E)}{\partial E} \right]_{E=E_F} \tag{3}$$

where $N(E)$ is the density of states and $\tau(E)$ is the relaxation time of conduction electrons [13]. In the present experiment, we can assume that the second term of equation (3) is field independent since the scattering centre does not differ through the transition [13]. Thus the sign change of S at each temperature might reflect the sign change of the energy derivative of the density of states at the Fermi level due to the Fermi-surface reconstruction associated with the transition between AF and F states.

The magnitude of S in Fe(3.5% Ni)Rh both in F and AF states is much larger than those in ordinary metals (e.g., about 14 and 0.4 μV K^{-1} for Fe and Rh, respectively, at 300 K) [13], suggesting a large value of $\mathrm{d}N(E)/\mathrm{d}E$ at the Fermi level according to equation (3). According to Wohlfarth, the first-order AF–F transition occurs in the itinerant-electron systems with large $\mathrm{d}N(E)/\mathrm{d}E$, large negative value of $\mathrm{d}^2N(E)/\mathrm{d}E^2$ or both around the Fermi level in the AF ground state [15]. The large and positive value of S in the AF state shows that at least the first condition predicted by Wohlfarth is satisfied for FeRh systems since the first term in equation (3) is dominant in the thermoelectric power as mentioned above.

The large value of S in the present FeRh system is comparable with that of $NaCo_2O_4$ which has been investigated as a new thermoelectric material recently [16]. The large S-value and the abrupt sign change of S may imply the possibility of application of FeRh systems as thermoelectric materials whose sign can be switched using the temperature and the magnetic field.

3.3. Specific heat

In order to investigate the field dependence of the density of states (DOS) at the Fermi level, we measured the temperature dependence of the specific heat (C) in the low-temperature region at $\mu_0 H = 0$, 2 and 7 T where the sample is in the AF, intermediate and F states, respectively. As shown in figure 7, C/T is linear against T^2 as usually observed for ordinary metals. The electronic specific heat coefficient γ was derived from the intercept of the C/T versus T^2 plot, which increases slightly for an applied field of 2 T and increases remarkably for an applied field of 7 T. The γ-values are estimated to be 6.0, 7.0 and 13.0 mJ mol^{-1} K^{-2} for $\mu_0 H = 0$, 2 and 7 T, respectively. The γ-values in the AF and F states are close to those reported in reference [5]. This fact suggests that the DOS at the Fermi level slightly increases through the transition from the AF to the intermediate state, and becomes about double in the F state. The small value of the DOS in the AF state implies the disappearance of some part of the Fermi surface due to formation of a superzone gap, leading to a large ρ. In the F state, the value of the DOS increases due to disappearance of the superzone gap, leading to a small ρ. This scenario supports the model where the reconstruction of the Fermi surface is the main origin of the GMR effect in FeRh systems.

In the C/T versus T^2 plot at $\mu_0 H = 0$ T, a small hump is observed around $T^2 = 10$ K^{-2}, suggesting another magnetic transition. We subtract $\gamma + \beta T^2$ assuming $\gamma = 6.0$ mJ mol^{-1} K^{-2} and $\beta = 0.142$ mJ mol^{-1} K^{-4} from the measured C/T, and plot the extra specific heat thus obtained (C_{ex}/T) against T as shown in figure 8. A small but clear peak in C_{ex}/T is observed around 2.8 K. A clear change is observed also in $\rho(T)$ at that temperature. These results indicate a new transition around 2.8 K. One possibility is that a long-range modulation of the

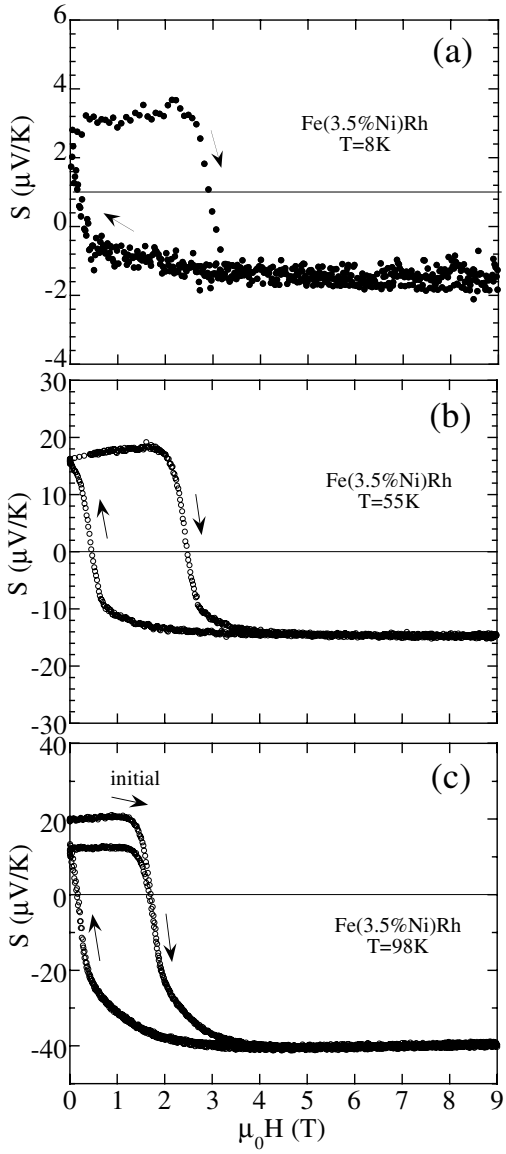

Figure 6. The field dependence of S at (a) 8, (b) 55 and (c) 98 K.

collinear AF magnetic (and crystalline) structure occurs, which modifies the AF superzone gap, leading to both a jump of $\rho(T)$ and a peak in C/T. Another possibility is that, even below T_0, AF ordering and the superzone gap formation are incomplete and AF ordering and the superzone gap formation proceed cooperatively at 2.8 K.

4. Discussion

In this section, we concentrate on the mechanism of GMR due to the reconstruction of the Fermi surface associated with the metamagnetic transition in the FeRh system. The remarkable

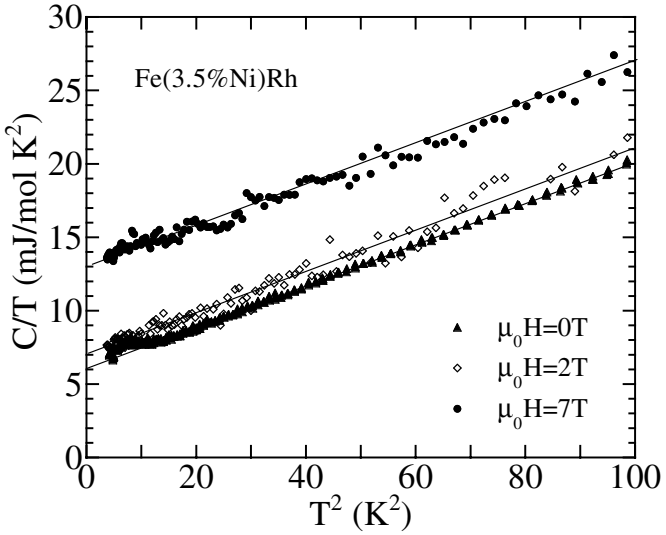

Figure 7. The temperature dependence of the specific heat, plotted as C/T versus T^2 at $\mu_0 H = 0$, 2 and 7 T.

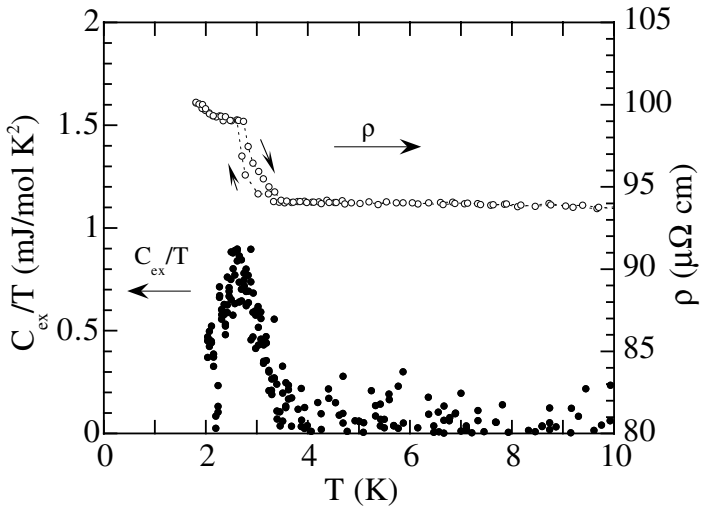

Figure 8. The temperature dependence of the extra part of the specific heat at $\mu_0 H = 0$ T, plotted as $C_{\rm ex}/T$ versus T, along with $\rho(T)$. $C_{\rm ex}/T$ was obtained by subtracting $\gamma + \beta T^2$ from the measured C/T assuming $\gamma = 6.0$ mJ mol^{-1} K^{-2} and $\beta = 0.142$ mJ mol^{-1} K^{-4}.

variations in the Hall effect, the thermoelectric power and the electronic specific heat coefficient with the reorientation of magnetic moments point to an appreciable change of the Fermi surface in FeRh systems at the metamagnetic transition. As the Fermi surface is intersected by new Brillouin-zone boundaries in the AF state, which leads generally to a gap in the electron dispersion relations, some part of the Fermi surface disappears; in other words, the carrier concentration reduces. This mechanism leads to an increase of ρ and a decrease of the γ-value (see figure 9(a)). However, the γ-value is only larger by a factor of two in the F state than that in

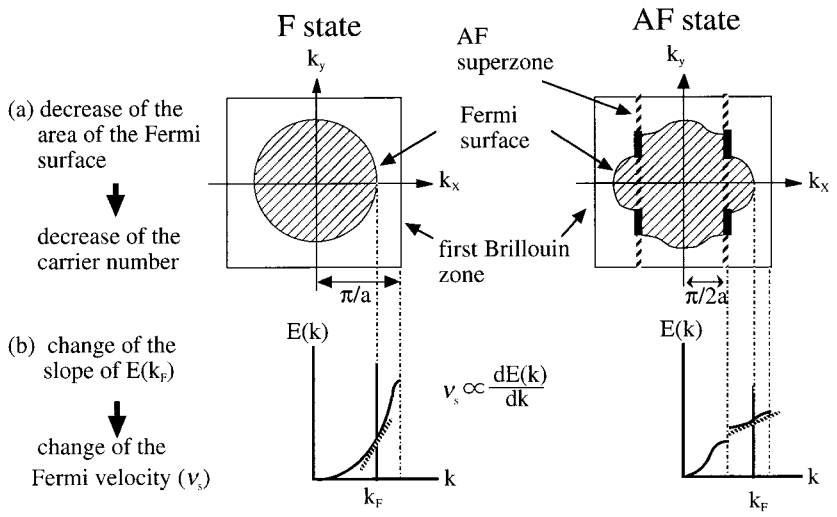

Figure 9. A schematic explanation of the mechanisms of (a) the reduction of the carrier concentration and (b) the reduction of the Fermi velocity (v_s) due to the reconstruction of the Fermi surface associated with the transition between the AF and the F state.

the AF state, which is not enough to explain the huge variation of ρ (by about a factor of eight) if the γ-value reflects only the area of the Fermi surface. We should take account of another mechanism to explain the GMR. One possible origin is the difference of the Fermi velocity resulting from the change of the band structure between the AF and F phases, which has been discussed by Oguchi to explain the GMR for magnetic multilayers [17]. According to a semi-classical theory of electrical conduction based on the Boltzmann equation, the resistivity is inversely proportional to the Fermi velocity v_s of the conduction electrons, which is given by the gradient of the energy dispersion in $\boldsymbol{k}$-space. Some of the band branches which cross the Fermi energy near the AF zone boundaries have a smaller gradient of the energy dispersion in the AF phase, leading to a smaller Fermi velocity (see figure 9(b)). Neglecting many-body mass enhancement, γ is described as

$$\gamma = \frac{\pi^2 k_B^2}{3} \frac{1}{8\pi^3 \hbar} \sum_s \int \frac{\mathrm{d}S_F}{|v_s|}. \tag{4}$$

According to equation (4), a smaller Fermi velocity leads to a larger γ-value. The two mechanisms of GMR due to the reconstruction of the Fermi surface—(a) reduction of the area of the Fermi surface and (b) the smaller Fermi velocity resulting from the superzone gap formation—both contribute to the enhancement of the resistivity for the AF state. In contrast, the two mechanisms contribute to γ oppositely: mechanism (a) decreases the γ-value and mechanism (b) enhances it. Thus we can understand the apparent discrepancy that ρ shows a huge change by about a factor of eight although γ changes by only a factor of two between the AF and F states.

5. Conclusions

We have investigated the magnetoresistance, magnetization, Hall effect, thermoelectric power and specific heat simultaneously for a Fe(3.5% Ni)Rh compound in order to investigate the origin of the GMR through the metamagnetic transition. The Hall coefficient changes its sign

through the metamagnetic transitions. The signs of the thermoelectric power are opposite for F and AF states. The electronic specific heat coefficient increases remarkably through the metamagnetic transition from the AF to the F state. These results indicate that the giant-magnetoresistance effect in FeRh compounds is predominantly caused by the reconstruction of the Fermi surface across the metamagnetic transition.

Acknowledgments

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