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Effect of scattering on intrinsic anomalous Hall effect investigated by Lorenz ratio

Y. Shiomi,[1,2] Y. Onose,[1,3] and Y. Tokura[1,2,3]
[1]*Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan*
[2]*Cross-Correlated Materials Research Group (CMRG), ASI, RIKEN, Wako 351-0198, Japan*
[3]*Multiferroics Project, ERATO, Japan Science and Technology Agency (JST), Tokyo 113-8656, Japan*
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We have investigated the effect of scattering on intrinsic anomalous Hall effect (AHE) for typical itinerant ferromagnets of Fe, Co, Ni, and their impurity-doped specimens in terms of the Hall Lorenz ratio $L_{xy} = \kappa_{xy}/\sigma_{xy}T$, the ratio of thermal ($\kappa_{xy}$) to electrical ($\sigma_{xy}$) Hall conductivities divided by temperature (T). We show that Lorenz ratio for the intrinsic anomalous Hall current (AHC) ($L_{xy}{}^{A}$) is almost constant and coincides with $L_0=2.44\times10^{-8}$ W Ω/K^2 (Wiedemann-Franz law) in the clean region (e.g., $\rho_{xx}=1-3\times10^{-6}$ Ω cm), indicating the scattering-free nature of intrinsic AHE. On the other hand, the Lorenz ratio for the AHC begins to decrease from $L_{xy}=L_0$ with increasing ρ_{xx} above $3-6\times10^{-6}$ Ω cm, which indicates the crossover from the scattering-free to scattering-dependent nature of AHE. The scattering rate ($\hbar/\tau$) corresponding to the crossover resistivity is comparable with the gap magnitude formed at the band anticrossing point caused by the spin-orbit interaction.

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I. INTRODUCTION

Recently, spin-dependent electronic transports have attracted renewed interests, because the spin current driven by electric field has unique potential beyond the conventional electronics, such as the dissipationless nature at room temperature. Anomalous Hall effect (AHE) is a prototypical example of the spin-related transport phenomena.[1] The AHE depends on magnetization in magnetic materials while the ordinary Hall effect is driven by the Lorentz force proportional to magnetic field. Because the dipole magnetic field generated by the magnetization is too small to explain the AHE, it has been believed from the early stage of the study that the AHE must be induced by the spin-orbit interaction. Several microscopic mechanisms relevant to the spin-orbit interaction have been proposed so far for the AHE. Some of the mechanisms are related to the scattering event under the influence of the spin-orbit interaction.[2,3] The skew scattering is induced by the change in the direction of the electron velocity in the course of scattering while the side jump by the position change. On the other hand, Karplus and Luttinger showed by the perturbation theory that the anomalous Hall current (AHC) can be induced by the external electric field and spin-orbit interaction irrespective of any scattering.[4] Recently, such an intrinsic mechanism of the AHE has been reinterpreted in terms of the Berry phase.[5–7] The Berry phase is the quantum phase induced by the adiabatic change in the parameter, on which the Hamiltonian depends. The intrinsic AHE is related to the Berry phase caused by the adiabatic change in momentum $\boldsymbol{k}$. The curvature of the Berry phase can be viewed as a fictitious magnetic field in $\boldsymbol{k}$ space. Microscopically, the Berry phase originates from the band mixture in terms of the spin-orbit interaction. In particular, the band anticrossing and gap formation by spin-orbit interaction give rise to the novel topological structure, namely, the magnetic monopole in $\boldsymbol{k}$ space, which works as a source of fictitious magnetic field.[6] While the longitudinal conductivity is determined by the electronic states on the Fermi surface, the anomalous Hall conductivity induced by the Berry phase is proportional to the sum of the fictitious magnetic fields, which are governed by the magnetic monopoles at the band anticrossing points. For this reason, the Berry-phase-induced AHC is protected by the gap structure at the band anticrossing point and hence robust against the scattering of electron to some extent.

The Berry-phase-induced AHC may provide an important clue to the development of low power-consumption device operating with dissipationless spin current. Conversely, the problem how robust the scattering-free nature of AHE against the increase in scattering is important; this is a motivation of the present investigation. As for the effect of scattering on the AHC, the σ_{xy} has been investigated with the variation in the scattering rate $1/\tau$ or the longitudinal conductivity σ_{xx} theoretically and experimentally.[1] Onoda *et al.*[8] theoretically studied the scattering-rate dependences of both the intrinsic and extrinsic AHEs and identified three characteristic regimes of the strength of the scattering rate: the skew-scattering-induced (extrinsic) AHE is rather dominant and $\sigma_{xy}\propto\sigma_{xx}$ in the clean limit of $\hbar/\tau < u_{\mathrm{imp}}E_{\mathrm{SO}}D$, where E_{SO}, u_{imp}, and D are the energies of the spin-orbit interaction, the impurity potential strength, and the density of states at the Fermi level, respectively. In the intermediate region ($u_{\mathrm{imp}}E_{\mathrm{SO}}D \leq \hbar/\tau \leq E_{\mathrm{F}}$), which is the most ubiquitous case in metals, the Berry-phase-induced intrinsic AHE is dominant and the Hall conductivity is rather independent of τ or σ_{xx}. This indicates that the intrinsic AHC is free from scattering. In the dirty limit ($\hbar/\tau \gg E_{\mathrm{F}}$), the Hall conductivity decreases with increasing scattering rate as $\sim\tau^{1.6}\propto\sigma_{xx}{}^{1.6}$ due to the broadening of the spectral width of quasiparticle. Experimentally, the relationship between σ_{xx} and σ_{xy} for AHE has been extensively investigated for more than five decades. In 1954, Kooi reported the anomalous Hall resistivity measured above 77 K satisfies the relation of $\rho_{yx}\propto\rho_{xx}{}^{2}$.[9] Lee *et al.*[10] confirmed the relation of $\rho_{yx}\propto\tau^{2}$ and ascribed it to the dissipationless nature of the intrinsic AHC for Br-doped $CuCr_2Se_4$. Recently, Miyasato *et al.*[11] investigated the longitudinal and Hall conductivities in several materials with a wide range of the resistivity and examined the relation between σ_{xx} and σ_{xy}. They observed that the crossover from the clean to the intermediate regime occurs around $\sigma_{xx} \sim 10^6\ \Omega^{-1}\,\text{cm}^{-1}$ and that of the intermediate to dirty one occurs around $\sigma_{xx} \sim 10^4\ \Omega^{-1}\,\text{cm}^{-1}$ for various transition-metal ferromagnets.

We have investigated the scattering effect on AHC in terms of thermal transport. Longitudinal thermal conductivity κ_{xx} is composed of electronic part (κ^{e}), phononic part (κ^{ph}), and magnonic part (κ^{mag}); $\kappa_{xx}=\kappa^{\text{e}}+\kappa^{\text{ph}}+\kappa^{\text{mag}}$. The Lorenz ratio ($L_{xx}{}^{\text{e}}$) can be defined as $L_{xx}{}^{\text{e}} \equiv \kappa^{\text{e}}/\sigma_{xx}T$, where σ_{xx} is the longitudinal electric conductivity. According to the Wiedemann-Franz law, the Lorenz ratio $L_{xx}{}^{\text{e}}$ coincides with the Lorenz number,

$$L_0 = \frac{\pi^2}{3}\left(\frac{k_{\text{B}}}{e}\right)^2 = 2.44 \times 10^{-8}\ \text{W}\,\Omega/\text{K}^2 \qquad (1)$$

at $T=0$. As temperature increases from $T=0$, $L_{xx}{}^{\text{e}}$ is known to decrease rapidly because the process of inelastic scattering suppresses the thermal current more effectively than the electrical current. Nevertheless, when the inelastic scattering caused by the interaction with phonon is dominated at elevated temperatures and the energy change in the course of the electron-phonon scattering becomes smaller than the thermal energy ($k_{\text{B}}T$), the Lorenz ratio again approaches L_0; this typically occurs in the high-temperature region above Debye temperature ($T>\Theta_{\text{D}}$). Thus, the Lorenz ratio is useful for studying the effect of the inelastic scattering of electron while it is difficult to estimate κ^{e} from the observed κ_{xx} alone. The latter problem can be overcome by investigating the Lorenz ratio for the Hall component; the Hall Lorenz ratio L_{xy} can be defined as $L_{xy}=\kappa_{xy}/\sigma_{xy}T$, where κ_{xy} is thermal Hall conductivity. The Wiedemann-Franz law also suggests $L_{xy}=L_0$ at $T=0$. Since phonon or magnon can hardly contribute to the thermal Hall conductivity, the Hall Lorenz ratio as defined by the observed σ_{xy} and κ_{xy} values can represent the genuine electronic contribution. The L_{xy} is expected to be sensitive to the inelastic scattering similarly to $L_{xx}{}^{\text{e}}$. Zhang *et al.*[12] reported that $L_{xy}/L_0 \simeq (L_{xx}{}^{\text{e}}/L_0)^2$, which is presumed by the τ^2 dependence of the normal Hall conductivity, holds good for Cu metal in a wide temperature region.

The Lorenz ratio for AHC ($L_{xy}{}^{\text{A}}$) was investigated for Ni and Cu-doped Ni.[13] We reported that $L_{xy}{}^{\text{A}}$ coincides with L_0 and shows minimal temperature dependence in the low-temperature region, indicating the scattering-free nature of AHE; the inelastic scattering does not affect the AHC. In a subsequent study,[14] we also observed that $L_{xy}{}^{\text{A}}$ for skew-scattering-induced AHE largely deviates from L_0 at finite temperature for Fe and its related alloys. The scattering-free nature of the Berry-phase (intrinsic) AHC and the dissipative feature of the skew-scattering-induced (extrinsic) one, as revealed by the Lorenz ratio study, correspond, respectively, to the intermediate ($\sigma_{xy}\sim$constant) and clean regimes ($\sigma_{xy}\sim\sigma_{xx}$) which were previously shown experimentally and theoretically.[8,11] Thus, the scattering effect on the intrinsic and extrinsic AHCs could be clarified by the measurement of the Hall Lorenz ratio. Then, it is meaningful to extend the Lorenz ratio study to examine how robust the scattering-free nature of the intrinsic AHC is against further increase in the scattering rate. For this purpose, we study here the anomalous part of the Lorenz ratio in the less-conductive region with increasing scattering rate. The electrical and thermal Hall conductivities are investigated in this work for Co, Ni, and Fe and their impurity-doped alloys (Cu-doped Ni, Co-doped Fe, Si-doped Fe, and Si-doped Co). We have found the $L_{xy}{}^{\text{A}}$ begins to deviate from L_0 when ρ_{xx} exceeds $3-6\times10^{-6}\ \Omega\,\text{cm}$. This crossover resistivity is much lower than that observed in the $\sigma_{xy}-\sigma_{xx}$ analysis[11] but the deduced scattering rate well corresponds to the energy scale of the gap induced by spin-orbit interaction around the band anti-crossing point.

II. EXPERIMENT

We used commercially available specimens for pure Co, Ni, and Fe. The alloys were made from pure elements by means of arc melting in an argon atmosphere. The longitudinal and Hall resistivities were investigated with use of physical property measurement system (Quantum Design Inc.). Measurements of longitudinal and transverse thermal conductivities were done with use of a steady-state method. We measured the longitudinal temperature gradient utilizing two thermometers (Cernox, Lake shore Cryotronics Inc.). As for the transverse temperature gradient, both type-E thermocouples ($T\gtrsim50$ K) and Cernox thermometers ($T<100$ K) were used. A resistance bridge (Model 370 AC Resistance Bridge, Lake shore Cryotronics Inc.) was used to measure the resistances of the thermometers. For the voltage measurement of thermocouples, we used the digital multimeter equipped with preamplifier (Keithley 2002, Keithley Instruments Inc.). A 1 kΩ chip resistance was attached to the end of the sample as a heat source.

III. RESULTS AND DISCUSSIONS

We show the temperature (T) dependence of ρ_{xx} for the pure and impurity-doped Ni, Fe, and Co in Fig. 1. The resistivities are very low at $T=0$ for the pure Ni, Co, and Fe ($\rho_{xx}\sim1-5\times10^{-7}\ \Omega\,\text{cm}$) and steeply increase with T due to phonon and spin-wave scatterings.[15] According to the former studies,[15] the resistivities of Fe, Co, and Ni in the low-T region is mainly attributed to the *s*-*s* and *s*-*d* scatterings by phonon and spin-wave and their contribution varies as T^5 or T^3 and as T^2, respectively. As T increases above Θ_{D}, the phonon scattering is much more important than the spin-wave scattering and the resistivity shows the T-linear dependence.[15] In fact, the T dependence of resistivity for the pure specimens changes from superlinear at the low T to linear at the high T, as seen in Figs. 1(a), 1(d), and 1(g). With doping impurity, the impurity scattering dominates the increasing residual resistivity. The resistivities of the most impurity-doped samples ($Ni_{0.9}Cu_{0.1}$, $Fe_{0.99}Si_{0.01}$, and $Co_{0.985}Si_{0.015}$) increase up to the order of $10^{-5}\ \Omega\,\text{cm}$ and show small and monotonous T dependences. The κ_{xx} values for all the samples are plotted as a function of T in Figs. 1(b),

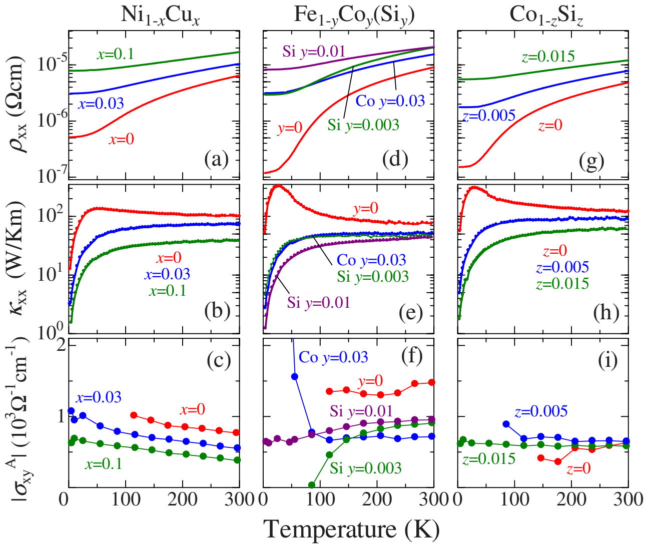

FIG. 1. (Color online) Temperature dependences of electrical resistivity (ρ_{xx}), thermal conductivity (κ_{xx}), and the anomalous parts of electrical Hall conductivities ($\sigma_{xy}{}^{A}$) for $Ni_{1-x}Cu_x$, $Fe_{1-y}Co_y(Si_y)$, and $Co_{1-z}Si_z$.

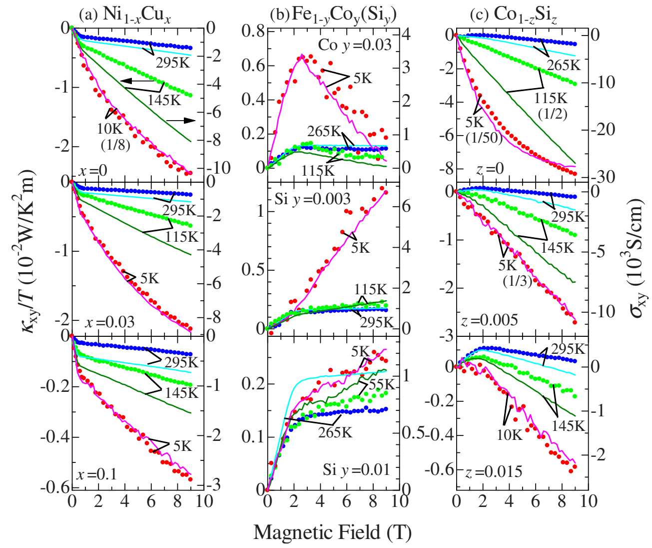

FIG. 2. (Color online) Magnetic field dependences of the thermal Hall conductivities divided by T (κ_{xy}/T, circles) and electrical Hall conductivities (σ_{xy}, solid lines) at various temperatures for (a) $Ni_{1-x}Cu_x$, (b) $Fe_{1-y}Co_y(Si_y)$, and (c) $Co_{1-z}Si_z$.

1(e), and 1(f). The electronic contribution is dominant in κ_{xx} for such good metals as the present samples. (Later, we will argue the relative proportion of electronic and phononic contributions in more detail in terms of the Lorenz ratio.) The κ_{xx} for pure specimens are large and show broad peaks around 30 K. This peak structure is owing to the crossover between the increase in κ_{xx} due to the small scattering rate and the decrease due to the small thermal energies of the electrons with lowering T. As the impurity concentration is increased, the κ_{xx} decreases reflecting the increase in ρ_{xx}, shows no peak, and increases monotonically with T for all the impurity-doped samples.

In Fig. 2, we show the magnetic field (H) dependence of σ_{xy} for all the samples with solid lines.[16] For $Ni_{0.9}Cu_{0.1}$, σ_{xy} is composed of the H-linear normal part and the M (magnetization)-linear anomalous part. The magnitude of the negative normal part increases with decreasing T. This is because $\sigma_{xy} \simeq \sigma_{xx}^2 \rho_{yx}$ and the σ_{xx} steeply increases with decreasing T. The anomalous part can be estimated from the linear extrapolation of the high-field data to zero magnetic field. The sign of the anomalous part is also negative but the magnitude does not show large T variation. When the Cu doping concentration is reduced, the T variation in the normal part becomes larger and the nonlinear field dependence emerges at low T, while the T dependence of the anomalous part remains small. The nonlinear behavior of the normal part is due to the large $\omega_c \tau$ (ω_c being cyclotron frequency) at the low-T region.[17] Similar T and doping dependences are seen in $Co_{1-x}Si_x$ system while the sign of the anomalous part is positive and the magnitude of the normal part is larger. For the case of Fe alloys, the normal part of σ_{xy} is quite small compared with the $Ni_{1-x}Cu_x$ and $Co_{1-x}Si_x$ systems and the anomalous part does not show large T variation in a high-T region. However, the H-linear and M-linear components change drastically in the low-T region below 100 K for $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$ owing to the skew scattering.[14] The nature of the skew scattering for the Fe alloys was discussed in a previous work;[14] we here concentrate on the high-T region of the Fe alloys above 100 K, where the intrinsic AHE is dominant.

In Figs. 1(c), 1(f), and 1(i), we show the T dependences of the anomalous Hall conductivities for all the samples. We could not accurately estimate the anomalous Hall conductivities in the low-T region ($T \lesssim 100$ K) for Fe, Co, Ni, and $Co_{0.995}Si_{0.005}$ since the anomalous Hall resistivities are too low because of high σ_{xx}. As shown in Figs. 1(c) and 1(i), the anomalous Hall conductivities for $Ni_{1-x}Cu_x$ and $Co_{1-x}Si_x$ do not show large T variation. The absolute values for $Ni_{1-x}Cu_x$ only slightly decrease with increasing T while those for $Co_{1-x}Si_x$ are almost constant [Fig. 1(i)]. These behaviors are consistent with the scattering-free nature of AHE. The anomalous Hall conductivities for $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$ are also nearly constant above 100 K, although a steep change below 100 K is caused by the skew scattering contribution.[14] For $Fe_{0.99}Si_{0.01}$, the anomalous Hall conductivities show small T dependence even in the low-T region and the skew scattering contribution ($\propto \sigma_{xx}$) are hardly discerned due to the increase in ρ_{xx}.

In Fig. 2, the measured values of κ_{xy}/T for all the samples are plotted with closed circles as a function of magnetic field in comparison with σ_{xy} (solid lines). The respective variations in κ_{xy}/T with the doping, field, and T share the qualitatively common features for all the samples; the field dependence of κ_{xy}/T at the lowest T coincides with that of σ_{xy} and the ratio is almost equal to L_0, satisfying the Wiedemann-Franz law. At high T, however, the magnitude of κ_{xy}/T is suppressed compared with σ_{xy} and the field dependences are different from each other. These are because the Lorenz ratio for AHC does not coincide with that for normal Hall current.

We define the Lorenz ratios for anomalous and normal Hall currents as follows:

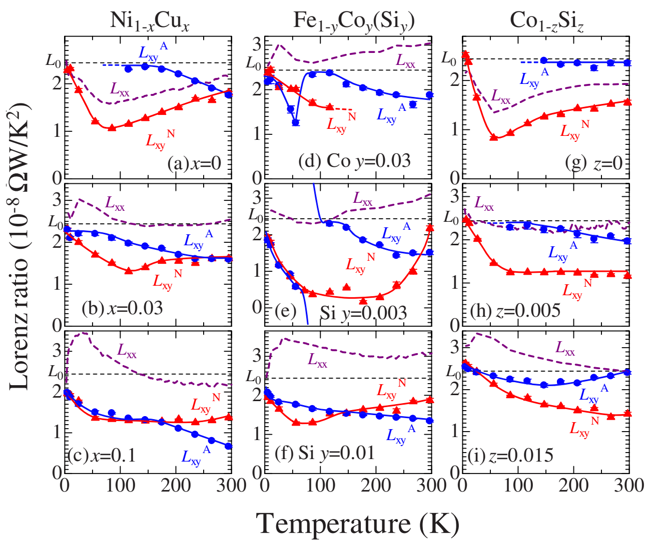

FIG. 3. (Color online) Temperature dependences of the Lorenz ratios for longitudinal conduction (L_{xx}, dashed lines), the normal Hall current ($L_{xy}{}^{\mathrm{N}}$, triangles), and the anomalous Hall current ($L_{xy}{}^{\mathrm{A}}$, circles). For $Fe_{0.97}Co_{0.03}$, $L_{xy}{}^{\mathrm{N}}$ is not shown above 120 K because the normal part of Hall resistivity is too small to accurately estimate.

$$L_{xy}{}^{\mathrm{A}} = \frac{\kappa_{xy}{}^{\mathrm{A}}}{\sigma_{xy}{}^{\mathrm{A}}T}, \tag{2}$$

$$L_{xy}{}^{\mathrm{N}} = \frac{\kappa_{xy}{}^{\mathrm{N}}}{\sigma_{xy}{}^{\mathrm{N}}T}, \tag{3}$$

where $\kappa_{xy}{}^{\mathrm{A}}$ and $\kappa_{xy}{}^{\mathrm{N}}$ ($\sigma_{xy}{}^{\mathrm{N}}$ and $\sigma_{xy}{}^{\mathrm{A}}$) are the anomalous and normal parts of thermal (electrical) Hall conductivity, respectively. We plot the T dependences of $L_{xy}{}^{\mathrm{N}}$ and $L_{xy}{}^{\mathrm{A}}$ together with the conventional Lorenz ratio for the longitudinal conductivity ($L_{xx}=\kappa_{xx}/\sigma_{xx}T$) in Fig. 3. Note that the L_{xx} differs, in principle, from $L_{xx}{}^{\mathrm{e}}(\equiv\kappa_{xx}{}^{\mathrm{e}}/\sigma_{xx}T)$ when the contributions of phonon (κ^{ph}) and magnon (κ^{mag}) to κ_{xx} are not neglected compared with the electronic one (κ^{e}). The L_{xx} for Ni and Co almost coincides with L_0 at the lowest T and becomes smaller than L_0 in the intermediate-T region. Then, it tends to recover to L_0 in the high-T region. This is same as the canonical T dependence of $L_{xx}{}^{\mathrm{e}}$ as explained in Sec. I, indicating that the electronic contribution is dominant in κ_{xx} in these highly conductive pure materials. For the heavily impurity-doped samples ($Ni_{0.9}Cu_{0.1}$, $Fe_{0.99}Si_{0.01}$, and $Co_{0.985}Si_{0.015}$), on the other hand, L_{xx} increases with increasing T from $T=0$ and shows a maximum around 30 K. The low-T behavior cannot be explained by the electronic contribution alone, implying some phonon contribution in these less-conducting samples.

The thermal Hall conductivity is composed of almost the electronic component alone since phonons or magnons can hardly show Hall effect.[18] The $L_{xy}{}^{\mathrm{N}}$ is expected to reflect the strength of the inelastic scattering similarly to $L_{xx}{}^{\mathrm{e}}$. As noted previously, the relation $L_{xy}{}^{\mathrm{N}}/L_0\propto(L_{xx}/L_0)^2$ is reported for Cu metal.[12] In the case of the pure Ni and Co, the T dependence of $L_{xy}{}^{\mathrm{N}}$ is similar to that of L_{xx}. To check the relation, we compare $L_{xy}{}^{\mathrm{N}}/L_0$ and $(L_{xx}/L_0)^2$ for pure Ni and Co in Fig. 4.

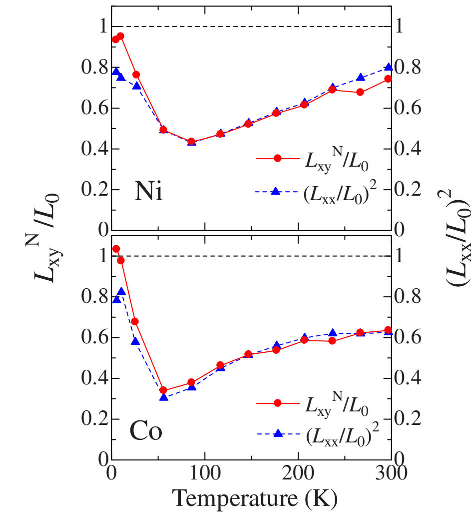

FIG. 4. (Color online) Comparison between $(L_{xx}/L_0)^2$ and $L_{xy}{}^{\mathrm{N}}/L_0$ for pure Ni and Co.

As is clear from the figure, the relation that $L_{xy}{}^{\mathrm{N}}/L_0 \propto(L_{xx}/L_0)^2$ holds also for ferromagnetic metals, Ni and Co. This relation ensures the usefulness of the Hall Lorenz ratio to investigate the effect of inelastic scattering. The $L_{xy}{}^{\mathrm{N}}$ for impurity-doped samples also decreases with increasing T from $T=0$, shows a minimum, and then tends to increase with T in the high-T region. For the Cu-doped Ni and Si-doped Co samples, the $L_{xy}{}^{\mathrm{N}}$ and the T at the minimum increase with increasing impurity concentration. This is because the ratio of inelastic to elastic scattering decreases with impurity doping. A similar impurity-concentration dependence of $L_{xx}{}^{\mathrm{e}}$ has been known for long.[20]

The T variation of $L_{xy}{}^{\mathrm{A}}$ is rather different from those of L_{xx} and $L_{xy}{}^{\mathrm{N}}$ as shown in Fig. 3. For pure Co, $L_{xy}{}^{\mathrm{A}}$ shows less deviation from L_0 above 140 K, while it cannot be accurately estimated below 140 K. This indicates that the AHC is hardly affected by the inelastic scattering. As discussed in a previous study on Ni,[13] this is an indication of scattering-free nature of Berry-phase-induced AHE. For $Co_{0.995}Si_{0.005}$, the $L_{xy}{}^{\mathrm{A}}$ coincides with L_0 and shows little T deviation in the low-T region, while it begins to decrease with increasing T from 150 K. With further increase in Si impurity ($Co_{0.985}Si_{0.015}$), $L_{xy}{}^{\mathrm{A}}$ decreases with T even in the low-T region. In the $Ni_{1-x}Cu_x$ system, the decrease in $L_{xy}{}^{\mathrm{A}}$ is observed around 160 K even for the pure ($x=0$) sample. This onset T of the decrease becomes 90 K for $x=0.03$. For $Ni_{0.9}Cu_{0.1}$ ($x=0.1$), the $L_{xy}{}^{\mathrm{A}}$ gradually decreases with increasing T from the lowest T. In the case of impurity-doped Fe samples, $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$, the $L_{xy}{}^{\mathrm{A}}$ shows steep change below 100 K, which is caused by the skew scattering as reported in the previous paper.[14] Apart from the influence of skew scattering, the $L_{xy}{}^{\mathrm{A}}$ coincides with L_0 around 100 K but decrease with T above 150 K. For $Fe_{0.99}Si_{0.01}$, the $L_{xy}{}^{\mathrm{A}}$ monotonically decreases with increasing T from the lowest T similarly to the high-doped cases of $Ni_{0.9}Cu_{0.1}$ and $Co_{0.985}Si_{0.015}$. Thus, there is the crossover from $L_{xy}{}^{\mathrm{A}}\sim L_0$ to $L_{xy}{}^{\mathrm{A}}<L_0$ in common for Ni-, Co-, and Fe-based systems. Since the decrease in $L_{xy}{}^{\mathrm{A}}$ is caused by the

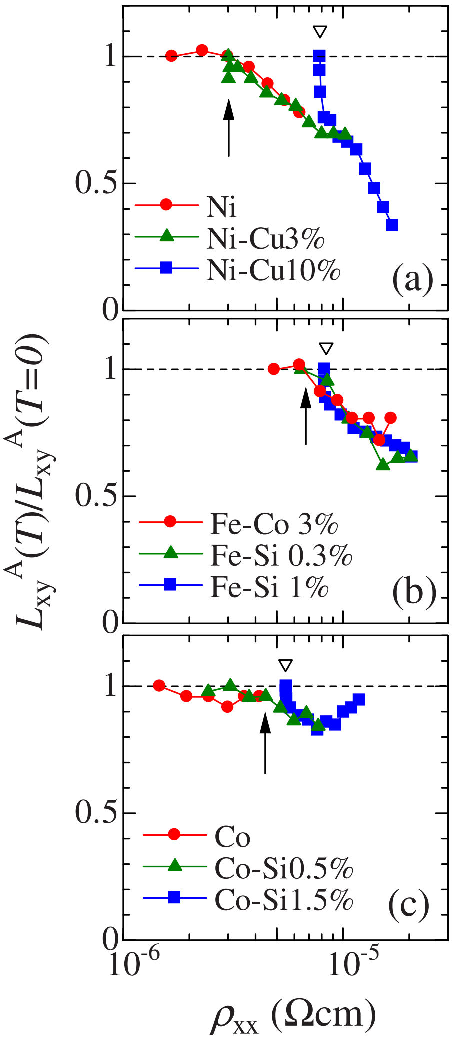

FIG. 5. (Color online) The ρ_{xx} dependence of $L_{xy}{}^{A}/L_0$ for (a) $Ni_{1-x}Cu_x$, (b) $Fe_{1-y}Co_y(Si_y)$, and (c) $Co_{1-z}Si_z$. The values of $L_{xy}{}^{A}(T=0)$ are estimated by extrapolation from the finite-T data in Fig. 3. We omit the low-T data governed by the skew scattering for $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$. The upward arrows indicate the critical ρ_{xx} value, above which $L_{xy}{}^{A}$ tends to deviate from its $T=0$ value ($\sim L_0$). (For open triangles, see text.)

effect of inelastic scattering, the scattering-free nature appears to be lost above the crossover T.

To discuss this phenomenon more quantitatively, we plot $L_{xy}{}^{A}$ as a function of the longitudinal resistivity ρ_{xx} in Fig. 5 for all the samples. The low-T data below 100 K for $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$ are omitted in this plot; they are dominated by the extrinsic skew scattering, which we do not discuss in this paper. For $Ni_{1-x}Cu_x$ system, the data except the low-T region for $x=0.1$ fall on to a single curve. The $L_{xy}{}^{A}$ is almost equal to L_0 in the low-ρ_{xx} region but it deviates in the high-ρ_{xx} region. Similar tendency is also observed for Co and Fe systems. Thus, the effect of inelastic scattering on the AHC seems to change at $\rho_{xx} \sim 3-6 \times 10^{-6}$ Ω cm for all the three systems. Incidentally, open triangles in Fig. 5 mark the sharp convergence of $L_{xy}{}^{A}(T)$ into $L_{xy}{}^{A}(T=0) \sim L_0$ in lowering T toward $T=0$ for the higher-resistive samples with higher doping; this should be distinguished from the above-mentioned crossover ρ_{xx} value. The crossover is not related to the energy gap of phonon nor magnon since the gaps should not be changed so drastically by the small amount of impurity doping. In addition, the crossover is dominated by the magnitude of resistivity value, which reflects the sum of the inelastic and elastic scattering rates, not the inelastic scattering alone. Therefore, the crossover should be caused by the scattering rates exceeding some critical energy scale. A similar type of crossover for the intrinsic AHE was proposed by Miyasato *et al.*[11] (experiment) and by Onoda *et al.*[8] (theory) as mentioned in Sec. I. In that case, the σ_{xx} dependence of σ_{xy} changes from $\sigma_{xy} \propto \sigma_{xx}{}^{0}$ to $\sigma_{xy} \propto \sigma_{xx}{}^{1.6}$ around $\rho_{xx} \sim 10^{-4}-10^{-3}$ Ω cm. However, this ρ_{xx} value is much larger than the present crossover value ($\rho_{xx} \sim 3-6 \times 10^{-6}$ Ω cm). (In the present case, any appreciable change in σ_{xy} is not discerned in Fig. 1 around our crossover value.) The Lorenz ratio study as presented here should be more sensitive to the scattering effect than the $\sigma_{xy}-\sigma_{xx}$ analysis and enabled us to identify another crossover of the AHE with change in scattering rate. Then, the next question is what is the origin of the crossover. From the observed value of the crossover resistivity, we can estimate the corresponding scattering rate, $\hbar/\tau \sim 20$ meV, assuming the carrier density $n \sim 1.7 \times 10^{22}/cm^3$ for Fe.[21] The energy scale is much smaller than Fermi energy (∼10 eV) and exchange splitting (∼1 eV). Only one quantity on a similar energy scale is the magnitude of energy gap at the band anticrossing point caused by the spin-orbit coupling; the gap of ∼70 meV is estimated in terms of first-principles band calculation for Fe.[7] The interband mixing is decisive for Berry-phase-induced AHE, therefore the broadening of the electronic spectrum greater than the gap magnitude around the band anticrossing point is likely to take away the scattering-free nature of AHC.

IV. CONCLUSIONS

In summary, we have investigated the scattering effect on the intrinsic AHC for Ni, Co, Fe, and their related alloys in terms of Hall Lorenz ratio, i.e., the ratio of thermal to electrical Hall conductivity divided by T; this quantity can measure the genuine electronic contribution to thermal current and the dissipation process of anomalous Hall conduction. The Lorenz ratio for the intrinsic AHC ($L_{xy}{}^{A}$) coincides with L_0 when ρ_{xx} is smaller than $3-6 \times 10^{-6}$ Ω cm. This is an indication of scattering-free nature of Berry-phase-induced intrinsic AHE. When ρ_{xx} exceeds the value of $3-6 \times 10^{-6}$ Ω cm, on the other hand, $L_{xy}{}^{A}$ decreases and deviates from L_0. The value of ρ_{xx} at the crossover corresponds to $\hbar/\tau \sim 20$ meV, which is comparable to the expected magnitude of the energy gap around band anticrossing point formed by the spin-orbit interaction.

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