Lorenz Number Determination of the Dissipationless Nature of the Anomalous Hall Effect in Itinerant Ferromagnets

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We have measured the thermal Hall conductivity for ferromagnetic Ni and $Ni_{0.97}Cu_{0.03}$. In the low temprature region ($\lesssim$100 K), we show for the first time that the Wiedemann-Franz law is satisfied even for the anomalous Hall current. While the Hall Lorenz number for the normal part decreases rapidly with temperature, that for the anomalous part shows much less deviation from the free-electron Lorenz number. This evidences the dissipationless nature of the anomalous Hall effect.

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The anomalous Hall effect (AHE) is the transverse conduction depending on the spontaneous magnetization and its direction in an itinerant ferromagnet. There appear to be several microscopic mechanisms to account for the AHE [1]. Some of the mechanisms are related to the scattering affected by the spin-orbit interaction. The deflected angle and the position change in the scattering process may depend on the direction of the spin of the scattered electron, owing to the spin-orbit interaction. These give rise to the skew scattering and side jump mechanisms, respectively [2,3]. On the other hand, Karplus and Luttinger showed by the perturbative description that in the presence of the spin-orbit interaction the interband matrix elements of the applied electric potential cause a finite velocity proportional to both the magnetization and electric field, which is denoted as anomalous velocity [4]. Recently, the anomalous velocity has been reinterpreted as induced by the Berry phase [1,5,6]. In general, the Berry phase is the quantum phase caused by the adiabatic change in some parameter space. The anomalous Hall conductivity is related to the Berry phase of the conduction electron in the momentum space. In contrast to the extrinsic mechanisms (skew scattering and side jump), such an intrinsic anomalous Hall effect as induced by the Berry phase is, in principle, not affected by the scattering. Experimentally, the relation that $\rho_{xy} \propto \rho_{xx}^2$ (namely, Hall conductivity $\sigma_{xy} \approx -\frac{\rho_{xy}}{\rho_{xx}^2}$ is independent of ρ_{xx}) is identified in some materials and is thought to be the evidence of the dissipationless nature of anomalous Hall current [7,8]. Nevertheless, the same ρ_{xx} dependence of ρ_{xy} is also expected for the side jump mechanism. In addition, the Hall conductivity may show the nontrivial temperature (T)- or doping-dependence because of its high sensitivity to the position of the chemical potential in the electronic band structure [9]. Another way to examine the origin and nature of the anomalous Hall effect is thus highly desired. Here, we take the approach to this problem in terms of the comparative study on the anomalous Hall currents of charge and heat in an itinerant ferromagnet.

The Wiedemann-Franz law represents the following relation between thermal conductivity (κ) and electric conductivity (σ), $\kappa = L_0 \sigma T$, where L_0 is the free-electron Lorenz number ($=2.44 \times 10^{-8}\ \Omega\mathrm{W/K^2}$) [10]. The law is satisfied only when the inelastic scattering is not present, viz., at $T = 0$. At a finite T, the Lorenz number ($L = \frac{\kappa}{\sigma T}$) becomes lower than L_0 because of the effect of inelastic scattering. This can be understood by considering the fact that the inelastic scattering process with a small change of wave vector affects the heat current but hardly the electric current. In the electron-phonon scattering case, the energy transfer is limited by the Debye temperature Θ. In the high T ($\gg \Theta$) region, the electron-phonon scattering can be viewed as the quasielastic scattering because the energy change in the course of the scattering is negligible compared with the temperature. Therefore, the Lorenz number recovers the original value L_0 in this limit. Thus, the Lorenz number of normal metal coincides with L_0 at $T = 0$ and high enough T, but is generally lower than L_0 in the middle range of T.

The thermal Hall effect (Righi-Leduc effect), i.e., the activation of the transverse thermal conductivity κ_{xy} by a magnetic field ($H \parallel z$), is a thermal analog of the Hall effect. While the longitudinal thermal conductivity is composed of both the electronic and the bosonic (phonon and magnon) parts, the charge-neutral boson can hardly contribute to the thermal Hall conductivity because of the absence of the Lorentz force [11]. The Hall Lorenz number L_{xy} is defined as $L_{xy} = \kappa_{xy}/\sigma_{xy}T$. According to the Wiedemann-Franz law, the L_{xy} is also equal to L_0 at $T = 0$, and may also deviate from L_0 at finite T. Zhang *et al.* [13] reported in the case of Cu that the L_{xy} shows a similar T dependence to $L(\equiv L_{xx})$. In an itinerant ferromagnet, the anomalous part of the thermal Hall effect is expected to show up. In fact, the spontaneous thermal Hall effect was already reported at several temperatures for Ni and Ni-Cu alloy, in old literature [14,15]. To the best of our knowledge, however, the Wiedemann-Franz law for the anomalous Hall current has never been reported, and the effect of

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inelastic scattering on the Hall Lorenz number is worth to investigate for itinerant ferromagnets from the contemporary view point of the anomalous velocity. In this work, we have measured the thermal Hall conductivity for Ni and $Ni_{0.97}Cu_{0.03}$, in which the anomalous Hall effect is anticipated to be induced by the intrinsic mechanism except for the low temperature (<100 K) region of Ni, as demonstrated by a recent study [16].

A commercially available pure Ni sample and a $Ni_{0.97}Cu_{0.03}$ sample prepared in this study by arc melting were used. The measurement of thermal conductivity was carried out by use of a conventional steady-state method. Two thermometers (CX-1050, Lakeshore Cryotronics, Inc.) were utilized to get the longitudinal T gradient. The transverse T gradient was measured using both the type E thermal couple ($T > 50$ K) and CX-1050 thermometers ($5\ \mathrm{K} < T < 100$ K). Longitudinal and Hall resistivities were measured in a Physical Property Measurement System (Quantum Design Inc.).

In Fig. 1(a), we show the T dependence of resistivity for Ni and $Ni_{0.97}Cu_{0.03}$. The resistivity for Ni is low ($\approx 0.5\ \mu\Omega$ cm) at the lowest T and increases rapidly with T. When Cu is doped into Ni, the residual resistivity is increased but the T dependence is almost unchanged. This indicates that only the elastic impurity scattering is increased in $Ni_{0.97}Cu_{0.03}$. The thermal conductivities for Ni and $Ni_{0.97}Cu_{0.03}$ are shown in Fig. 1(b). The thermal conductivity for Ni increases gradually with decreasing T from 300 K, and shows a peak around 50 K. On the other hand, the thermal conductivity for $Ni_{0.97}Cu_{0.03}$ monotonically decreases with decreasing T. The nominal Lorenz number $L\ (= \frac{\kappa}{\sigma T})$ does not differ too much from L_0 in a whole T region for both the samples ($1.6 \times 10^{-8} \leq L \leq 3.2 \times 10^{-8}$), indicating that the electronic contribution is dominant to the thermal conductivity.

Figures 1(d) and 1(e) show the H dependence of the Hall resistivity for Ni and $Ni_{0.97}Cu_{0.03}$. The Hall resistivity for Ni is composed of the normal part ($\propto H$) and the anomalous part ($\propto M$) at 297 K. While the normal part shows little T dependence above 86 K, the anomalous Hall resistivity decreases largely with decreasing T. At the lowest T, the anomalous Hall resistivity almost vanishes, and the normal part shows nonlinear field dependence. When the Cu is doped into Ni, the anomalous part becomes larger, and can be observed even at the lowest T, where the nonlinear normal part is observed as well. The Hall resistivities for Ni and $Ni_{0.97}Cu_{0.03}$ are similar to those reported previouly [17,18]. The nonlinear behavior for the normal part may be related to a large value of $\omega_c\tau$ (ω_c and τ being cyclotron frequency and relaxation time, respectively). The anomalous part of Hall resistivity can be obtained by the linear extrapolation from the high field above the saturation field of the magnetization ($\approx$0.6 T) to zero field. Using this procedure, we obtain and show in Fig. 1(c) the T dependence of the anomalous part of the Hall conductivity for Ni and $Ni_{0.97}Cu_{0.03}$. The anomalous Hall conductivity for Ni increases in the absolute magnitude gradually with decreasing T. Below 86 K, it is difficult to estimate it because of the small anomalous Hall resistivity. The anomalous Hall conductivity for $Ni_{0.97}Cu_{0.03}$ shows a similar behavior, but can be estimated even at the lowest T.

In Figs. 2(a) and 2(b), we show the H dependence of the thermal Hall conductivity κ_{xy} for Ni at various T. At 297 K, the thermal Hall conductivity is composed of the M-linear anomalous part and the H-linear normal part. Similarly to the normal part of electrical Hall conductivity,

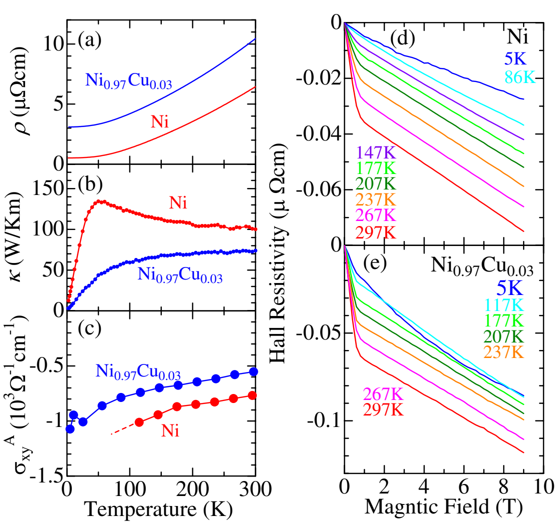

FIG. 1 (color online). (a)–(c) Temperature dependence of (a) resistivity, (b) thermal conductivity, and (c) anomalous part of Hall conductivity for Ni and $Ni_{0.97}Cu_{0.03}$. (d),(e) Magnetic field dependence of Hall resistivity at various temperatures for (d) Ni and (e) $Ni_{0.97}Cu_{0.03}$.

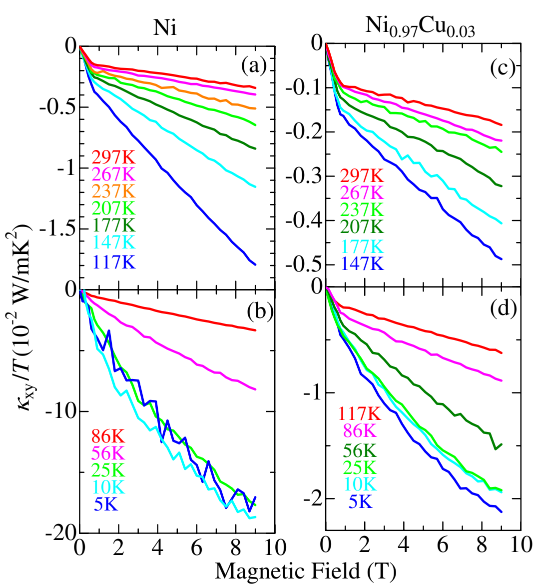

FIG. 2 (color online). Magnetic field dependence of thermal Hall conductivity at various temperatures for (a),(b) Ni and (c),(d) $Ni_{0.97}Cu_{0.03}$.

which varies with T as $\sim 1/\rho_{xx}(T)^2$, the normal part of the κ_{xy}/T increases largely with decreasing T. On the other hand, the anomalous part of the κ_{xy}/T does not show appreciable T variation, which is also similar to the electrical case. Below 86 K, it is difficult to identify the anomalous component, and the normal part shows a nonlinear behavior with H. The thermal Hall conductivity for $Ni_{0.97}Cu_{0.03}$ shows similar T and H dependences [Figs. 2(c) and 2(d)]. Compared with the pure Ni case, the normal component is smaller, while the anomalous one almost unchanged. The thermal Hall conductivity shows a kink around 0.6 T at 5 K, indicating the observable anomalous component even at the lowest T. This enables us to discuss the T dependence of the anomalous thermal Hall conductivity in the whole T region for $Ni_{0.97}Cu_{0.03}$.

Figure 3 compares the H dependences of thermal and electrical conductivities at various T for Ni and $Ni_{0.97}Cu_{0.03}$. In this figure, the ordinate scales for the respective conductivities are determined so that the anomalous components for the both conductivities coincide with each other. For Ni, the whole field dependence of thermal Hall conductivity is fully scaled with that of the electrical one at 297 K. Below 207 K, however, the normal component of the thermal conductivity is suppressed compared with that of electrical conductivity. Below 56 K, the anomalous components of the both conductivities can hardly be discerned, but the field dependences become scaled again with each other. A similar T variation is seen for $Ni_{0.97}Cu_{0.03}$. The difference is that the anomalous component for $Ni_{0.97}Cu_{0.03}$ can be discerned down to the lowest T, where the both conductivities are scaled well. The violation of the scaling of the field dependences in the middle T range implies that the anomalous component of thermal Hall conductivity needs separate consideration from that of normal component.

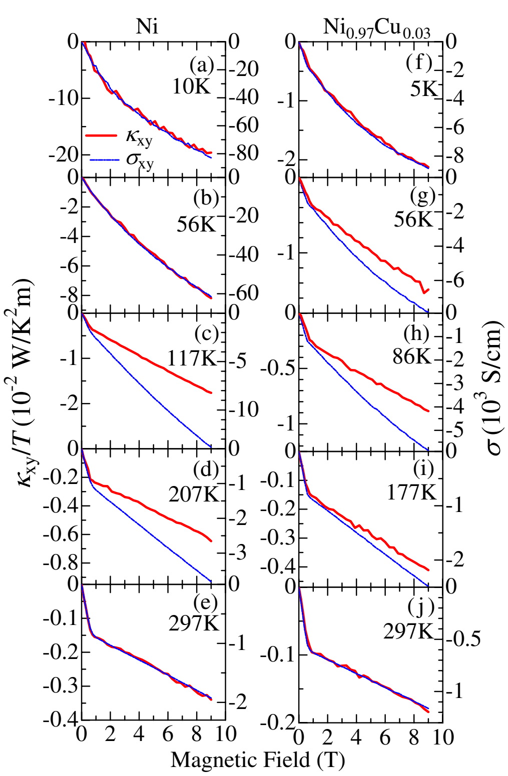

FIG. 3 (color online). Magnetic field dependence of both electrical and thermal Hall conductivities at respective temperatures for (a)–(e) Ni and (f)–(j) $Ni_{0.97}Cu_{0.03}$.

To discuss the anomalous and normal parts separately, we define the Lorenz numbers for the anomalous and normal components, respectively, as $L_{xy}^A = \kappa_{xy}^A/\sigma_{xy}^A T$ and $L_{xy}^N = \kappa_{xy}^N/\sigma_{xy}^N T$, where κ_{xy}^A, κ_{xy}^N, σ_{xy}^A, and σ_{xy}^N are the anomalous and normal components of thermal and electrical Hall conductivities, respectively. The L_{xy}^A and L_{xy}^N for Ni and $Ni_{0.97}Cu_{0.03}$ are shown in Figs. 4(a) and 4(b). The L_{xy}^N for Ni at the lowest T almost coincides with L_0. The L_{xy}^N decreases steeply with T and shows a minimum around 100 K. Above 100 K, the L_{xy}^N gradually increases with T. For $Ni_{0.97}Cu_{0.03}$, L_{xy}^N shows a similar behavior, but the decrease of L_{xy}^N is less significant. These behaviors are similar to the longitudinal part of Lorenz number (L_{xx}) for a normal metal as well as to the L_{xy} observed for a Pauli-paramagnetic metal Cu, and can be explained by the effect of inelastic scattering [10,13], as discussed later. The L_{xy}^A for both Ni and $Ni_{0.97}Cu_{0.03}$ almost coincides with L_0 in the low T region. This suggests that the Wiedemann-Franz law is valid also for the anomalous Hall current. For Ni, the L_{xy}^A remains almost constant below 180 K, while it is difficult to estimate the L_{xy}^A below 90 K. Above 180 K, the L_{xy}^A gradually decreases with T. The L_{xy}^A can be ob-

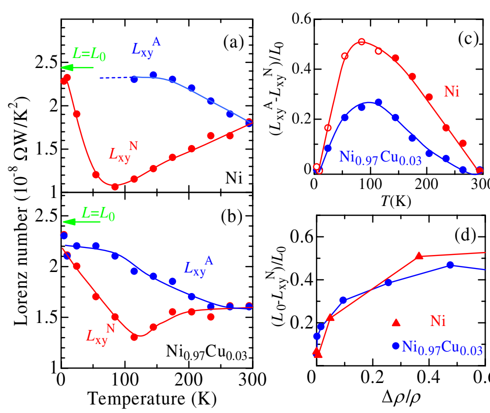

FIG. 4 (color online). (a),(b) Temperature dependences of the Hall Lorenz numbers for the anomalous (L_{xy}^A) and normal parts (L_{xy}^N) for (a) Ni and (b) $Ni_{0.97}Cu_{0.03}$. (c) Difference between L_{xy}^A and L_{xy}^N for Ni and $Ni_{0.97}Cu_{0.03}$ normalized by the free-electron Lorenz number L_0. (d) The decrease of the L_{xy}^N normalized by L_0 plotted against $\Delta\rho/\rho_0$, where ρ_0 and $\Delta\rho$ are residual resistivity and $\rho - \rho_0$, respectively. The arrows in (a) and (b) indicate the free-electron Lorenz number L_0. The solid lines are merely the guide to the eyes. The open red circles in (c) are plotted by assuming that the L_{xy}^A for Ni is constant below 100 K, as represented by the dashed line in (a).

tained over the whole T region for $Ni_{0.97}Cu_{0.03}$. The L_{xy}^A remains almost constant below 90 K down to the lowest T and decreases gradually with increasing T from 90 K. Thus, the L_{xy}^A shows little T dependence in the low T region for both Ni and $Ni_{0.97}Cu_{0.03}$. In the high T region, the L_{xy}^A decreases gradually, but the decrease is much less than that of L_{xy}^N. The differences between L_{xy}^A and L_{xy}^N for Ni and $Ni_{0.97}Cu_{0.03}$ are shown explicitly in Fig. 4(c). For the both samples, the L_{xy}^A and L_{xy}^N are almost equal to each other at the lowest T and room T, while the difference is significant around 100 K.

If there were no inelastic scattering, both the L_{xy}^A and L_{xy}^N should be equal to L_0. In other words, the deviation of the Lorenz number from L_0 signals the magnitude of the effect of inelastic scattering. In Fig. 4(d), the relative deviation of the L_{xy}^N from L_0, $(L_0 - L_{xy}^N)/L_0$, in the low T region is plotted against $\Delta\rho/\rho_0$, where ρ_0 is the residual resistivity and $\Delta\rho = \rho - \rho_0$. Since ρ_0 and $\Delta\rho$ are caused by the elastic impurity scattering and inelastic scattering, respectively, the $\Delta\rho/\rho_0$ stands for the magnitude of inelastic scattering relative to that of the elastic scattering. The scaling behavior of the L_{xy}^N suggests that the decrease of Hall Lorenz number for the normal component certainly signals the contribution of the inelastic scattering. The reason why the decrease of L_{xy}^N for Ni is more significant than that for $Ni_{0.97}Cu_{0.03}$ is that the effect of inelastic scattering is relatively weak for $Ni_{0.97}Cu_{0.03}$ due to the larger ρ_0. Considering that L_{xy}^A is close to L_0 in the low temperature region, where the scaling shown in Fig. 4(d) holds good, the T-dependent difference between L_{xy}^A and L_{xy}^N can be also ascribed to the influence of the inelastic scattering. When the intrinsic mechanism works for the anomalous Hall effect in these samples, the transverse current is anticipated to be almost free from the effect of inelastic scattering, and is considered as "dissipationless current". Recently, the detailed theoretical [19] and experimental [16] works have shown to what extent the intrinsic anomalous Hall current can be viewed as dissipation-free; the result is that even for the intrinsic mechanism the anomalous Hall conductivity depends on the scattering rate to some extent, but the effect is minimal in the range of $E_{so} < \hbar/\tau < E_F$ (1–$10\ \mu\Omega\,\text{cm} \leq \rho \leq 10^{-3}\ \Omega\,\text{cm}$), where E_{so} and E_F are the energy of spin-orbit coupling and Fermi energy, respectively. In the present case, the scattering rate is well within this range, except for the low T region ($T < 100$ K) of Ni. The T variation of Hall conductivity is weak in spite of the large residual resistivity ratio ($\rho_{300\text{ K}}/\rho_0 \approx 12$ and 3 for Ni and $Ni_{0.97}Cu_{0.03}$, respectively). The T independence of L_{xy}^A in the low T region indicates the *dissipationless* nature, namely, the anomalous Hall current should be hardly affected by the inelastic scattering nor by the elastic scattering in this regime. Incidently, a pure dissiaptionless heat current would not affect the lattice temperature. However, the accumulated thermal energy should be eventually released to the lattice system, which we believe was measured by the actual experiment.

As noted above, the anomalous Hall conductivity weakly depends on the scattering rate due to the broadening of spectral function even for the intrinsic mechanism. The gradual decrease of the L_{xy}^A in the high T region might be due to this weak influence of the scattering, although the effect of inelastic scattering on the Hall Lorenz number cannot reach the level of the quantitative understanding as yet because of lack in the elaborate theory on thermal Hall conductivity.

In conclusion, we have observed the anomalous thermal Hall effect for Ni and $Ni_{0.97}Cu_{0.03}$. The Hall Lorenz number for the anomalous component L_{xy}^A of both Ni and $Ni_{0.97}Cu_{0.03}$ almost coincides with the free-electron Lorenz number L_0 in the low temperature T region. This confirms the validity of Wiedemann-Franz law for the anomalous Hall current as well. While the Hall Lorenz number for the normal component L_{xy}^N decreases significantly with increasing T up to around 100 K, the L_{xy}^A for the both samples shows little T dependence in that low T region. This is an indication of the dissipationless nature of the intrinsic anomalous Hall effect.

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