PHYSICAL REVIEW B **79**, 100404(R) (2009)

Extrinsic anomalous Hall effect in charge and heat transport in pure iron, $Fe_{0.997}Si_{0.003}$, and $Fe_{0.97}Co_{0.03}$

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(Received 26 January 2009; published 16 March 2009)

We have investigated thermal and electrical Hall conductivities for nominally pure and impurity-doped iron to examine the skew-scattering-induced anomalous Hall effect in the low-temperature clean-limit regime. In Fe, $Fe_{0.997}Si_{0.003}$, and $Fe_{0.97}Co_{0.03}$, we have observed the steep change in the electrical and thermal Hall conductivities below 100 K. The abrupt change in the Lorenz numbers for the anomalous Hall component (L_{xy}^A) that includes even the sign change signaling the opposite flow of charge and heat currents is also observed around the crossover temperature of Hall conductivity. These observations indicate the emergence of the dissipative anomalous Hall current induced by the skew scattering below 100 K.

DOI: 10.1103/PhysRevB.79.100404 PACS number(s): 75.47.−m, 75.50.Bb, 72.15.Eb, 72.20.My

Among many important transport phenomena related to electronic spins, spin Hall effect (SHE) and anomalous Hall effect (AHE) are of renewed great interest.[1] The SHE is the emergence of transverse spin current upon the application of longitudinal electric field. In ferromagnets, the up and down spin population is unbalanced and the charge current also flows transversely, which is denoted as AHE. There are several mechanisms proposed for the AHE and SHE. Mechanisms of skew scattering and side jump are relevant to the scattering affected by the spin-orbit interaction.[2–4] The skew scattering is caused by the direction change in the scattered electron, while the side jump by the position change. On the other hand, Karplus and Luttinger[5] described the perturbation approach; that even without any scattering the longitudinal electrical field can induce the transverse velocity of electrons owing to the effect of spin-orbit interaction on the Bloch electron. Recently, such an intrinsic mechanism irrelevant to the scattering has been theoretically reinterpreted as induced by the Berry phase of the Bloch electron in $\boldsymbol{k}$ space.[6–8] In general, the adiabatic change in parameter gives rise to the Berry phase when Hamiltonian depends on the parameter. Since the energy of Bloch state is determined by the momentum $\boldsymbol{k}$, the band electrons acquire the Berry phase in the $\boldsymbol{k}$ space. The Berry phase theory for AHE suggests that the Hall conductivity is proportional to the integral of the Berry phase curvature over the occupied states.

While recent theoretical and experimental works show that the AHE can be well explained by the Berry phase theory in most cases, Onoda *et al.*[9] theoretically showed that the skew-scattering mechanism should be dominant in the clean-limit regime. This is because the Hall conductivity caused by the skew scattering increases linearly with the mean-free path while the Hall conductivity due to Berry phase is independent of scattering rate. Miyasato *et al.*[10] investigated the AHE for nominally pure Fe to explore the clean-limit regime. They observed some crossover in this regime and assigned it to the intrinsic to extrinsic crossover of AHE. This kind of crossover behavior of AHE was already reported in literature of decades ago.[11,12] Nevertheless, the extrinsic skew-scattering-induced AHE could hardly be analyzed quantitatively because of the difficulty in discriminating it from the magnetic-field-nonlinear normal Hall conductivity in this region.[10] The purpose of the present study is to identify the skew-scattering-induced AHE and examine its nature in terms of the Lorenz number for Hall conductivity.

The key to distinguish both mechanisms of AHE is to see the dissipation effect; the intrinsic AHE independent of scattering events should be dissipationless in nature, while the extrinsic one should show more or less the energy dissipation at finite temperatures. The dissipation effect in charge transport phenomena can be probed with use of the Lorenz number L.[13] The L is the ratio of thermal (κ) to electrical conductivity (σ) divided by temperature (T); namely, $L=\kappa/\sigma T$. At $T=0$, the Lorenz number coincides with the constant $L_0 = 2.44\times10^{-8}$ WΩ/K^2 prescribed for free-electron system, exactly obeying the Wiedemann-Franz law. As the temperature is increased, the Lorenz number usually decreases since the thermal current is more effectively suppressed than the electrical current by inelastic scatterings. This is because the inelastic scattering even with a tiny change in momentum affects the heat current but hardly the electric current. Therefore, the deviation of L from L_0 measures the influence of the inelastic scattering on the electronic transport. The Lorenz number for Hall conductivity,[14] defined as $L_{xy}=\kappa_{xy}/\sigma_{xy}T$, can genuinely probe the electronic transport (no phonon contribution). In this Rapid Communication, we have studied the electrical and thermal Hall conductivities for nominally pure and impurity-doped Fe to argue the intrinsic vs extrinsic features of AHE in terms of Lorenz number.

We used a commercially available specimen of Fe (Nilaco Co., 4*N* purity; major impurity is Si of 16 ppm). The intentionally impurity-doped specimens were made by arc melting in argon atmosphere. These specimens were cut into a rectangular parallelepiped shape with a typical size of $4\times1\times0.1$ mm^3. Longitudinal and transverse (Hall) resistivities were measured in a physical property measurement system (Quantum Design, Inc.). The thermal conductivity was measured with use of a steady-state method. Two thermometers (CX-1050, Lakeshore Inc.) were utilized to measure the longitudinal temperature gradient. The transverse tempera-

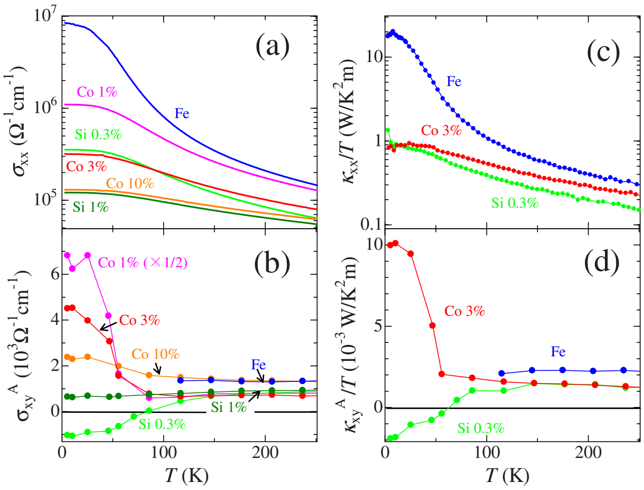

FIG. 1. (Color online) Longitudinal and anomalous Hall conductivities of charge and heat. Temperature (T) profiles of (a) the electrical conductivity (σ_{xx}), (b) the anomalous part of electrical Hall conductivity (σ_{xy}^A), (c) the thermal conductivity (κ_{xx}) divided by T, and (d) the anomalous part of thermal Hall conductivity (κ_{xy}^A) divided by T.

ture gradient was measured using type E thermocouples ($T>40$ K) and thermometers (5 K$<T<$80 K).

Figure 1(a) shows the temperature dependence of the electrical conductivity σ_{xx} for nominally pure Fe, $Fe_{0.997}Si_{0.003}$, $Fe_{0.99}Si_{0.01}$, $Fe_{0.99}Co_{0.01}$, $Fe_{0.97}Co_{0.03}$, and $Fe_{0.9}Co_{0.1}$. The σ_{xx} at the lowest temperature (σ_0) for nominally pure Fe is as large as 10^7 Ω^{-1} cm^{-1} but decreased by doping Co or Si. In Fig. 1(c), we show the temperature variations of the thermal conductivity κ_{xx} for nominally pure Fe, $Fe_{0.97}Co_{0.03}$, and $Fe_{0.997}Si_{0.003}$. The magnetic-field dependences of Hall resistivities, electric ρ_{yx} and thermal w_{yx}, at 5 K are shown in Fig. 2. While the Hall resistivity for nominally pure Fe shows quadratic field dependence, those for $Fe_{0.997}Si_{0.003}$, $Fe_{0.99}Si_{0.01}$, and $Fe_{0.97}Co_{0.03}$ are composed of well-defined H-linear normal and M-linear anomalous terms. It is to be noted that the anomalous term is negative for $Fe_{0.997}Si_{0.003}$ contrary to other specimens, signaling the sensitivity of the low-temperature Hall resistivity to the species and amounts of the impurities[15] and hence to the impurity scattering.

The anomalous parts of the Hall conductivities σ_{xy}^A and κ_{xy}^A/T for all the samples are plotted as a function of temperature in Figs. 1(b) and 1(d). These are calculated from the linear extrapolation from the high-field values above the saturation field of the magnetization, as exemplified by dashed lines in Fig. 2. The σ_{xy}^A and κ_{xy}^A/T for nominally pure Fe cannot be deduced below 100 K because of difficulty in estimating the anomalous part due to the high-field nonlinear behavior of ρ_{yx} and $w_{yx}T$. The σ_{xy}^A and κ_{xy}^A/T for all the samples are almost constant above 100 K, reminiscent of the dominant intrinsic component both for the charge and heat Hall transports. The difference in the σ_{xy}^A at high temperatures ($>$100 K) among the samples may be ascribed to the difference in Fermi energy which is dependent on the species and amount of the dopant. For Si-doped Fe the σ_{xy}^A and κ_{xy}^A/T decrease with decreasing temperature below 100 K. For Co-doped Fe, by contrast, the σ_{xy}^A and κ_{xy}^A/T increase with decreasing temperature below 100 K. We plotted the low-temperature change of σ_{xy}^A, $\sigma_{xy}^s=\sigma_{xy}^A(T=5\text{ K})-\sigma_{xy}^A(T=207\text{ K})$, as a function of the longitudinal conductivity σ_{xx} at the lowest temperature (5 K) in the inset of Fig. 2. As the impurity content increases, the magnitude of σ_{xy}^s decreases rapidly and the intrinsic AHE seems to become dominant. The σ_{xy}^s is proportional to σ_{xx} with the positive and negative slopes for Co-doped and Si-doped samples, respectively. The linear relation agrees with the skew-scattering theory,[2,9] and the sign and slope should reflect the effective impurity potential. The onset temperature of the skew scattering is almost identical ($\sim$100 K). Thus, the contribution of the skew scattering is also clearly observed in κ_{xy}^A/T below 100 K. As described below, the Hall Lorenz number $L_{xy}=\kappa_{xy}/\sigma_{xy}T$ at the lowest temperature (5 K), where the skew scattering totally dominates the Hall effect, is $\sim L_0$.

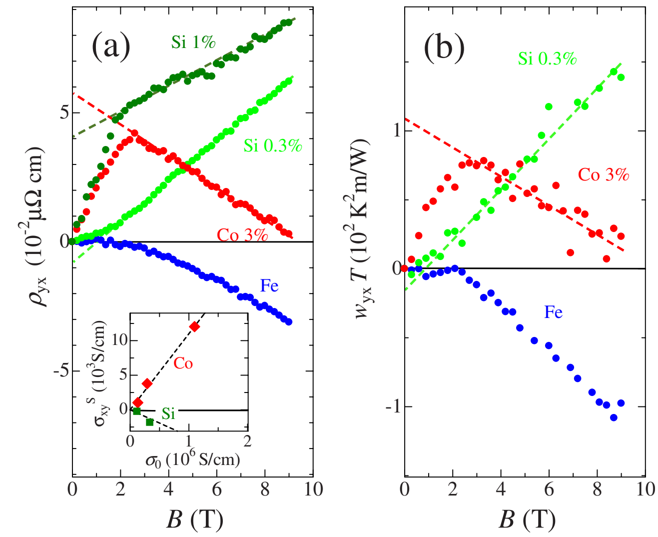

FIG. 2. (Color online) Electrical and thermal Hall resistivities. (a) and (b) Magnetic-field (B) dependence of the electrical and thermal Hall resistivity [$\rho_{yx}=\sigma_{xy}/(\sigma_{xx}^2+\sigma_{xy}^2)$ and $w_{yx}=\kappa_{xy}/(\kappa_{xx}^2+\kappa_{xy}^2)$, respectively] at $T=5$ K for each sample. The dashed lines exemplify the extrapolation procedure for the estimate of the anomalous Hall resistivity. The inset shows the skew-scattering-induced anomalous Hall conductivity (σ_{xy}^s) (see text for definition) plotted against the lowest-temperature conductivity (σ_0, inverse of residual resistivity) at 5 K. Diamonds (red online) and squares (green online) represent the data of Co-doped and Si-doped samples, respectively.

Figure 3 compares the thermal and electrical Hall conductivities at various temperatures for these samples. At any temperature, the electrical and thermal Hall conductivities (σ_{xy} and κ_{xy}) for $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$ are composed of the M-linear anomalous part and the H-linear normal part, apart from the indistinguishable feature for nominally pure Fe below 100 K. The field dependence of κ_{xy} roughly scales with that of σ_{xy} for all the samples. Nevertheless, the ratio of the normal to anomalous components is different between κ_{xy} and σ_{xy} in some cases (e.g., at 117 K for Fe and 86 K for $Fe_{0.997}Si_{0.003}$). This signals that the Lorenz number for anomalous component $L_{xy}^A=\kappa_{xy}^A/\sigma_{xy}^A T$ is different from that for normal component $L_{xy}^N=\kappa_{xy}^N/\sigma_{xy}^N T$.[13] On the basis of these

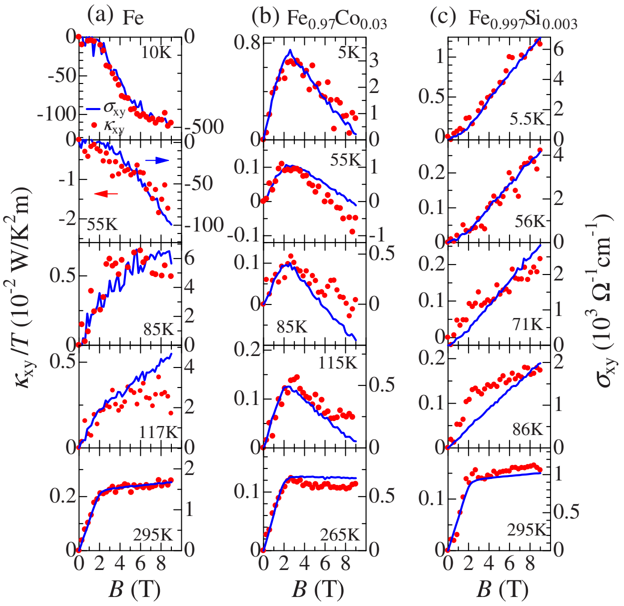

FIG. 3. (Color online) Comparison between electrical and thermal Hall conductivities. Magnetic-field (B) dependence of electrical and thermal Hall conductivities (σ_{xy} solid lines and κ_{xy}/T dots, respectively) for (a) nominally pure Fe, (b) $Fe_{0.97}Co_{0.03}$, and (c) $Fe_{0.997}Si_{0.003}$.

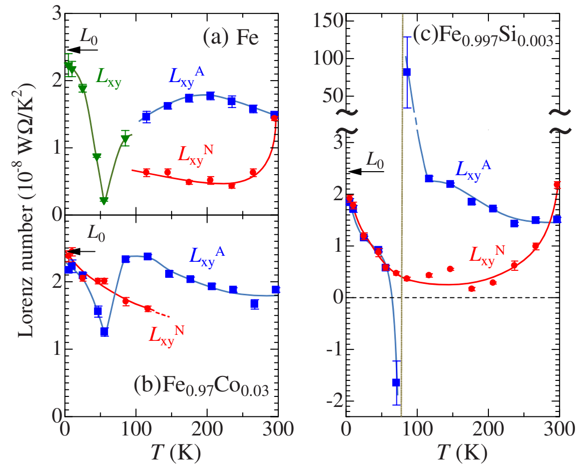

FIG. 4. (Color online) Lorenz numbers for normal and anomalous Hall currents. Temperature (T) dependence of Lorenz number for Hall current for (a) nominally pure Fe, (b) $Fe_{0.97}Co_{0.03}$, and (c) $Fe_{0.997}Si_{0.003}$. The squares (L_{xy}^A; blue online) and circles (L_{xy}^N; red online) indicate the Lorenz numbers for anomalous and normal Hall currents (see text for definition), respectively. Triangles (green online) in (a) stand for the Lorenz number determined by the scaling of the whole magnetic-field dependence of thermal and electrical conductivities. The solid and dashed lines are merely the guide for the eyes. L_0 ($=2.44\times10^{-8}$ $\mathrm{W\Omega/K^2}$) is the free electron value prescribed by the Wiedemann-Franz law.

data, we show L_{xy}^A and L_{xy}^N in Fig. 4. For $Fe_{0.97}Co_{0.03}$, we cannot obtain L_{xy}^N accurately above 120 K because of the small magnitude of the normal Hall conductivity. Although the Hall conductivity for nominally pure Fe below 100 K cannot be decomposed into normal and anomalous parts, the scaling of the whole field dependence of both the Hall conductivities is fairly good in this temperature region. We plot in Fig. 4(a) this Lorenz number L_{xy} for nominally pure Fe. The L_{xy}^A for $Fe_{0.97}Co_{0.03}$ [Fig. 4(b)] increases gradually with decreasing temperature from 300 K; almost coincides with L_0 around 150 K, but abruptly decreases below 80 K. After showing a sharp minimum around 60 K, it recovers to L_0 at the lowest temperature. The L_{xy}^A for $Fe_{0.997}Si_{0.003}$ [Fig. 4(c)] varies with temperature similarly to the case of $Fe_{0.97}Co_{0.03}$ above 100 K. Below 100 K, however, it divergently increases and then turns to a negative value at 71 K. Below 50 K, it again increases toward L_0 at the lowest temperature. These steep changes in L_{xy}^A below 100 K are not discerned for the higher-doped sample $Fe_{0.99}Si_{0.01}$ (not shown). The L_{xy}^N for all the samples shows a large downward deviation from L_0 with a broad minimum around 150 K. The total L_{xy} (with indistinguishable normal and anomalous components) for nominally pure Fe shows a sharp minimum around 60 K similar to the L_{xy}^A for $Fe_{0.97}Co_{0.03}$.

The effect of inelastic scattering as measured by the deviation of Lorenz number from L_0 can be seen not only in L_{xy}^N but also in the low-temperature (<100 K) L_{xy}^A. Around 150 K, the L_{xy}^A for all the samples is larger than L_{xy}^N and close to L_0. This indicates that the AHC is hardly affected by the inelastic scattering in this temperature range; this is due to the dissipationless nature of the Berry-phase-induced AHE.[13] The abrupt change in L_{xy}^A for $Fe_{0.97}Co_{0.03}$ and $Fe_{0.997}Si_{0.003}$ occurs around the onset temperature (~100 K) of the skew scattering in the electrical conduction. The decrease in L_{xy}^A for $Fe_{0.97}Co_{0.03}$ can be thus ascribed to the nondissipative to dissipative crossovers of AHE due to the emergence of skew scattering at finite temperatures. The negative Lorenz number for $Fe_{0.997}Si_{0.003}$ at 71 K means that the direction of the electrical current is opposite to that of the thermal current. This surprising phenomenon can be explained as follows. The Lorenz number can be defined for both the Berry-phase- and the skew-scattering-induced AHCs, L_{xy}^B and L_{xy}^s. The L_{xy}^A is then expressed as

$$L_{xy}^A = \frac{\kappa_{xy}^s + \kappa_{xy}^B}{\sigma_{xy}^s + \sigma_{xy}^B}\frac{1}{T} = \frac{L_{xy}^s\sigma_{xy}^s + L_{xy}^B\sigma_{xy}^B}{\sigma_{xy}^s + \sigma_{xy}^B}. \qquad (1)$$

Here, κ_{xy}^s, κ_{xy}^B, σ_{xy}^s, and σ_{xy}^B stand for the thermal and electrical Hall conductivities induced by the skew scattering (extrinsic) and the Berry phase (intrinsic), respectively. L_{xy}^s is quite temperature dependent, whereas L_{xy}^B is nearly constant ($\sim L_0$) because of the dissipationless nature of the intrinsic AHE. Note that the sign of σ_{xy}^s and κ_{xy}^s is opposite to that of σ_{xy}^B and κ_{xy}^B for $Fe_{0.997}Si_{0.003}$. This is the reason why a negative value is observed for L_{xy}^A of $Fe_{0.997}Si_{0.003}$ in the crossover temperature region. Conversely, the *negative* Lorenz number for $Fe_{0.997}Si_{0.003}$ is viewed as the compelling evidence for the existence of two distinct AHCs, i.e., induced by the skew scattering and the Berry phase.

In conclusion, we have successfully identified the skew-scattering contribution to the electrical Hall conductivity (σ_{xy}^s) as well as to the thermal one (κ_{xy}^s) for iron metals with controlled doping of impurity. The Lorenz number for the AHC is larger than that for the normal one and close to the value (L_0) prescribed by the Wiedemann-Franz law in the high-temperature region where the Berry-phase-induced intrinsic AHC is dominant. In the low-temperature region below 100 K, by contrast, the Lorenz number for AHC is much smaller than L_0 or even negative. These features firmly evidence the emergence of the dissipative Hall current induced by the skew scattering in the low- (<100 K) but finite temperature region, in addition to the persistent dissipationless intrinsic anomalous Hall current.

Valuable discussions with N. Nagaosa and S. Onoda are gratefully acknowledged. This work was in part supported by Grants-In-Aid for Scientific Research from the MEXT (Contracts No. 19684011, No. 20340086, and No. 20046004).

[1] N. Nagaosa, J. Phys. Soc. Jpn. **75**, 042001 (2006).
[2] J. Smit, Physica (Amsterdam) **24**, 39 (1958).
[3] L. Berger, Phys. Rev. B **2**, 4559 (1970).
[4] A. Crépieux and P. Bruno, Phys. Rev. B **64**, 014416 (2001).
[5] R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
[6] T. Jungwirth, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. **88**, 207208 (2002).
[7] Z. Fang, N. Nagaosa, K. S. Takahashi, A. Asamitsu, R. Mathieu, T. Ogasawara, H. Yamada, M. Kawasaki, Y. Tokura, and K. Terakura, Science **302**, 92 (2003).
[8] Y. Yao, L. Kleinman, A. H. MacDonald, J. Sinova, T. Jungwirth, D. S. Wang, E. Wang, and Q. Niu, Phys. Rev. Lett. **92**, 037204 (2004).
[9] S. Onoda, N. Sugimoto, and N. Nagaosa, Phys. Rev. Lett. **97**, 126602 (2006); Phys. Rev. B **77**, 165103 (2008).
[10] T. Miyasato, N. Abe, T. Fujii, A. Asamitsu, S. Onoda, Y. Onose, N. Nagaosa, and Y. Tokura, Phys. Rev. Lett. **99**, 086602 (2007).
[11] P. N. Dheer, Phys. Rev. **156**, 637 (1967).
[12] A. K. Majumdar and L. Berger, Phys. Rev. B **7**, 4203 (1973).
[13] Y. Onose, Y. Shiomi, and Y. Tokura, Phys. Rev. Lett. **100**, 016601 (2008).
[14] Y. Zhang, N. P. Ong, Z. A. Xu, K. Krishana, R. Gagnon, and L. Taillefer, Phys. Rev. Lett. **84**, 2219 (2000).
[15] The H-linear "normal term" seems to be also strongly affected by the impurity scattering. This cannot be explained by the conventional Hall effect induced by Lorenz force. In this regime, the normal and anomalous components may be mixed with each other.