Anomalous Hall Heat Current and Nernst Effect in the $CuCr_2Se_{4-x}Br_x$ Ferromagnet

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In a ferromagnet, an anomalous Hall heat current, given by the off-diagonal Peltier term α_{xy}, accompanies the anomalous Hall current. By combining Nernst, thermopower, and Hall experiments, we have measured how α_{xy} varies with hole density and lifetime τ in $CuCr_2Se_{4-x}Br_x$. At low temperatures T, we find that α_{xy} is independent of τ, consistent with anomalous-velocity theories. Its magnitude is fixed by a microscopic geometric area $\mathcal{A} \sim 34$ Å^2. Our results are incompatible with some models of the Nernst effect in ferromagnets.

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In a ferromagnet, the anomalous Hall effect (AHE) is the appearance of a spontaneous Hall current flowing parallel to $\mathbf{E} \times \mathbf{M}$, where $\mathbf{E}$ is the electric field and $\mathbf{M}$ the magnetization [1]. Karplus and Luttinger (KL) [2] proposed that the AHE current originates from an anomalous-velocity term which is nonvanishing in a ferromagnet. The topological nature of the KL theory has been of considerable interest recently [3–6]. Experimentally, strong evidence for the dissipationless nature of the AHE current has been obtained in the spinel ferromagnet $CuCr_2Se_{4-x}Br_x$. Lee *et al.* [7] reported that, despite a 1000-fold increase in the resistivity ρ induced by varying the Br content x, the anomalous Hall conductivity (normalized per carrier and measured at 5 K) stays at the same value, in agreement with the KL prediction. A test of the anomalous-velocity theory against the AHE in Fe has also been reported [8].

It has long been known that an anomalous heat current density $\mathbf{J}^Q$ also accompanies the AHE current in the absence of any temperature gradient [9,10]. In principle, $\mathbf{J}^Q$ can provide further information on the origin of the AHE, but almost nothing is known about its properties. A weak heat current is a challenge to measure. Instead, one often performs the "reciprocal" Nernst experiment in which a temperature gradient $-\nabla T$ produces a transverse charge current, which is detected as a Nernst electric field $\mathbf{E}_N$ parallel to $\mathbf{M} \times (-\nabla T)$. However, in previous Nernst experiments on ferromagnets [9–11], J^Q was not found because other transport quantities were not measured. Combining the Nernst signal with the AHE resistivity ρ'_{xy} and the thermopower, we have determined how the transport quantity α_{xy} relevant to J^Q varies in $CuCr_2Se_{4-x}Br_x$ as the hole density n_h and carrier lifetime τ are greatly changed under doping. We show that α_{xy} has a strikingly simple form, with its magnitude scaled by a microscopic geometric area $\mathcal{A}$.

We apply a gradient $-\nabla T \| \hat{\mathbf{x}}$ to an electrically isolated sample in a magnetic field $\mathbf{H} \| \hat{\mathbf{z}}$. Along $\hat{\mathbf{x}}$, the charge current driven by $-\nabla T$ is balanced by a backflow current produced by a large E_x which is detected as the thermopower $S = E_x/|\nabla T|$. Along the transverse direction $\hat{\mathbf{y}}$, however, both E_x and $-\nabla T$ generate Hall-type currents. In general, the charge current in the presence of $\mathbf{E}$ and $-\nabla T$ is $\mathbf{J} = \boldsymbol{\sigma} \cdot \mathbf{E} + \boldsymbol{\alpha} \cdot (-\nabla T)$, with $\boldsymbol{\sigma}$ and $\boldsymbol{\alpha}$ the electrical and thermoelectric ("Peltier") conductivity tensors, respectively. Setting $J_y = 0$, we obtain the Nernst signal $e_N \equiv E_y/|\nabla T| = \rho\alpha_{xy} + \rho_{xy}\alpha$, where $\alpha \equiv \alpha_{xx}$ [12]. Hence, as noted, the Nernst signal results from the two distinct y-axis charge currents, $\alpha_{yx}(-\nabla T)$ and $\sigma_{yx}E_x$. In a ferromagnet, the former is our desired gradient-driven current, whereas the latter comprises the "dissipationless" AHE current and the weak ordinary Hall current.

In terms of the thermopower $S = \rho\alpha$ and Hall angle $\tan\theta_H = \rho_{yx}/\rho$, we may express α_{xy} as

$$\rho\alpha_{xy} = e_N + S\tan\theta_H. \tag{1}$$

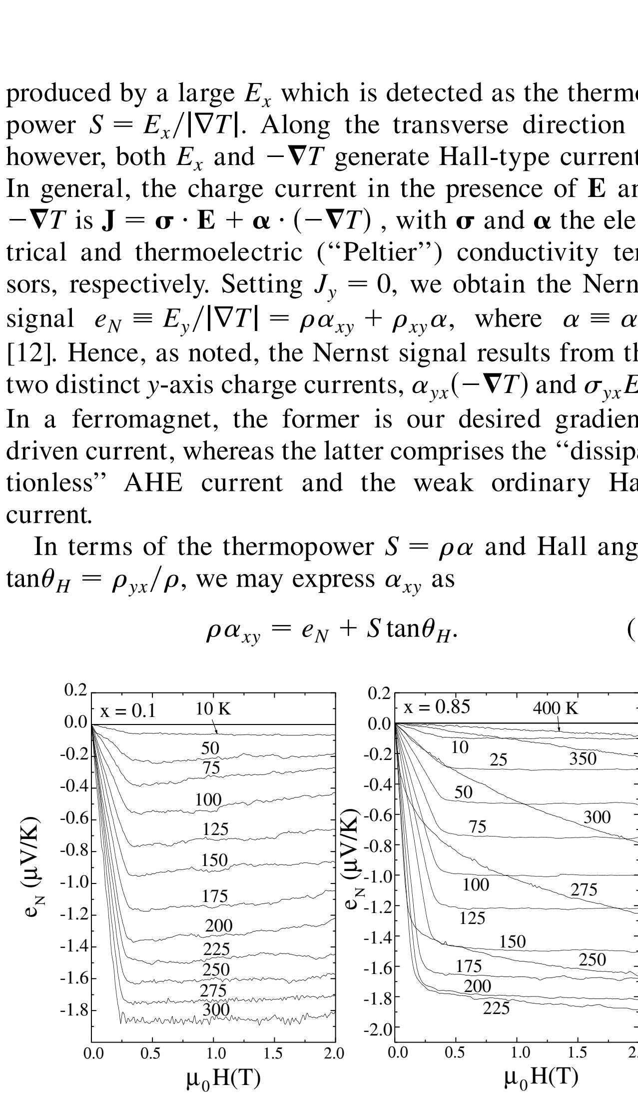

FIG. 1. Curves of the measured $e_N = E_y/|\nabla T|$ versus H in $CuCr_2Se_{4-x}Br_x$, with $x = 0.1$ (left panel) and 0.85 (right panel). In the ferromagnetic state below T_C, e_N saturates to a constant when H exceeds H_s reflecting the M-H curve. The scaling factor Q_s increases rapidly as T increases from 10 K to T_C. In the right panel, e_N continues to scale as the M-H curve in the paramagnetic regime (275–400 K).

Hence, to find α_{xy}, we need to measure e_N, S, ρ_{xy}, and ρ. Knowing α_{xy}, we readily find the transverse heat current $J_y^Q = \tilde{\alpha}_{yx} E_x$, since $\tilde{\alpha}_{yx} = \alpha_{yx} T$ by Onsager reciprocity.

The spinel $CuCr_2Se_4$ is a conducting ferromagnet with a Curie temperature $T_C \sim 450$ K. Because the exchange between local moments in Cr is mediated by superexchange through 90° Cr-Se-Cr bonds rather than the carriers, T_C is not significantly reduced even when the hole population n_h drops by a factor of 30 under Br doping (M at 5 K actually increases by 20%) [7,13]. Using iodine vapor transport, we have grown crystals with x from 0.0 to 1.0. As x increases from 0 to 1, the value of ρ at 5 K increases by $\sim 10^3$, while ρ'_{xy}/n_h increases by $\sim 10^6$ [7]. The tunability of n_h and the robustness of M under doping make this system attractive for studying charge transport in a lattice with broken time-reversal symmetry. The behavior of ρ, M, and ρ'_{xy} versus x are described in Ref. [7].

Figure 1 shows profiles of e_N versus H at selected T in two samples with $x = 0.1$ and 0.85 and $T_C = 400$ and 275 K, respectively. As noted above, $e_N(T, H)$ is the sum of two terms, both of which scale as M. The magnitude $|e_N|$ initially increases as H rotates domains into alignment and then saturates to a constant for $H > H_s$, the saturation field. The sign of e_N—negative in all samples—reflects the sign of the dominant term [14].

In the sample with $x = 0.85$, the curves above T_C show that the scaling also holds in the paramagnetic regime where the susceptibility has the Curie-Weiss form $\chi \sim 1/(T - T_C)$ in weak H. In analogy with the Hall resistivity $\rho_{xy} = R_0 \mu_0 H + R_s \mu_0 M$, with R_0 and R_s the ordinary and anomalous Hall coefficients, respectively, it is customary to express the scaling between the e_N-H and M-H curves by writing

$$e_N = Q_0 \mu_0 H + Q_s \mu_0 M. \tag{2}$$

For $T < T_C$ in all samples, the Q_0 term cannot be resolved, so that $e_N \simeq Q_s \mu_0 M$. Moreover, below 50 K, M changes only weakly with x (by 20% over the whole doping range), so that the saturated value of the Nernst signal e_N^{sat} differs from Q_s by a factor that is only weakly x dependent.

The Nernst signal has very different characteristic behaviors below and above T_C. As an example, Fig. 2 shows e_N^{sat} measured at 2 T in the sample with $x = 1.0$ ($T_C = 210$ K). Between 5 and 100 K, e_N^{sat} increases linearly with T. Above 100 K, e_N^{sat} rises more steeply to a sharp peak 200 K, and then falls steeply above T_C. As noted, in the paramagnetic regime, the Nernst signal matches the behavior of M as a function of both T and H. Figure 2 shows that the T dependence of e_N closely follows that of $M = \chi H$ (both are measured at 2 T). The experiment shows that, in a gradient, fluctuations of the paramagnetic magnetization lead to a significant transverse electrical current that is proportional to the average magnetization (this has not been noted before, to our knowledge). We express the proportionality as

$$\alpha_{xy} = \beta M \qquad (T > T_C), \tag{3}$$

where β is only weakly T dependent (it decreases by 5% between 250 and 400 K). The parameter β plays the important role of relating the magnitudes of the paramagnetic M and the transverse electronic current (through the Nernst signal). Its minuscule value ($\beta \simeq 2 \times 10^{-7}\ \mathrm{K}^{-1}$ at 250 K) reflects the strikingly weak coupling between the fluctuating M and e_N in a

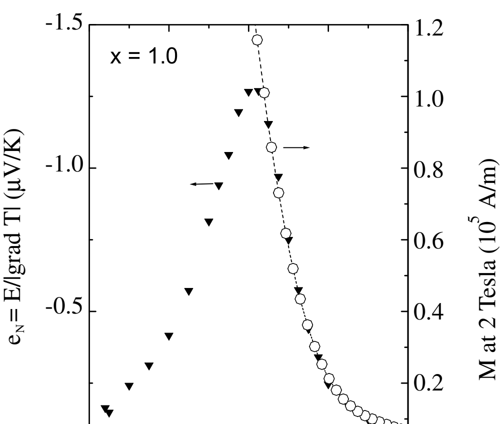

FIG. 2. The T dependence of the Nernst signal e_N (solid triangles) measured at 2 T in the sample with $x = 1.0$. Above T_C, e_N is compared with the paramagnetic magnetization M at 2 T (open circles).

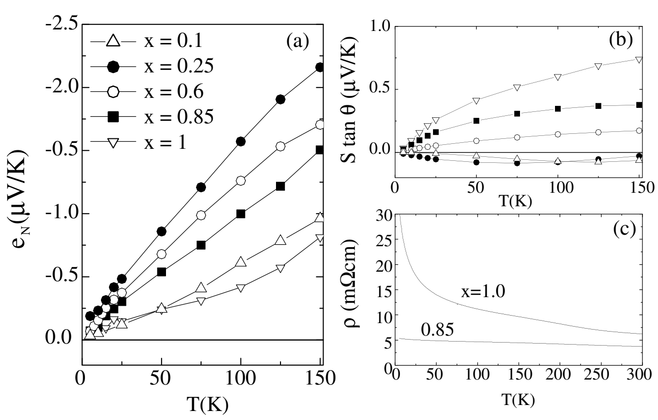

FIG. 3. (a) Curves of e_N versus T below 150 K in five samples with doping $0.1 \le x \le 1.0$ showing nominal T-linear behavior at low T ($H = 2$ T). The slopes vary nonmonotonically with x. (b) shows the Hall-current term $S \tan\theta_H$ measured in the same samples at $H = 2T$. For $x > 0.3$, $S \tan\theta_H$ is opposite in sign from e_N [the symbol key applies to both (a) and (b)]. (c) shows the sharp change in the ρ-T profiles in the samples with $x =$ 0.85 and 1.0 ($H = 0$). At low T, ρ at 0.85 is metallic, but at 1.0 ρ reveals hopping between strongly localized states. See Ref. [7] for ρ versus T for $x < 0.85$.

ferromagnet; a sizeable $M \sim 10^5$ A/m produces a Nernst signal of only $\sim 2\ \mu$V/K. With the growth of long-range magnetic order below T_C, Eq. (3) ceases to be valid.

In the ferromagnetic state, we restrict our attention to the regime below 100 K, where $e_N^{\rm sat}$ is nominally linear in T. Figure 3(a) shows curves in this regime for the five samples studied. The slopes of the low-T curves are not monotonic in x. As x is increased from 0.1, the slope attains a maximum value at $x = 0.25$, and then decreases to a value close to its initial value when x reaches 1.0. This is perhaps not surprising since e_N involves transport quantities S, ρ, and ρ_{xy} with opposite trends versus x. By Eq. (1), we may find the curve of α_{xy} versus T by adding the curves e_N and $S\tan\theta_H$ [Fig. 3(b)] and dividing by ρ. The data in Figs. 3(a) and 3(b) show that these terms are opposite in sign for $x > 0.3$. With increasing x, their mutual cancellation suppresses α_{xy} strongly. In particular, at the largest x (0.85 and 1.0), the cancellation is nearly complete and α_{xy} is very small below 100 K; i.e., the observed e_N is nearly entirely from the AHE of the backflow current. For $0.1 \leq x < 0.3$, $S\tan\theta_H$ is negligible and e_N largely reflects the behavior of α_{xy}. We exclude from our study the undoped compound $CuCr_2Se_4$ because e_N and ρ'_{xy} were not resolved at low T. These trends emphasize the importance of knowing all four transport quantities, instead of just e_N, to discuss $\mathbf{J}^Q$ meaningfully.

Finally, the derived curves of α_{xy} versus T are shown in Fig. 4. In contrast to the nonmonotonic behavior of e_N versus x, α_{xy} varies linearly with T as $\alpha_{xy} = b(x)T + c$, where the slope $b(x)$ now decreases monotonically as x increases from 0.1 to 1.0 (Fig. 4). In all samples except $x = 0.25$, the parameter c—probably extrinsic in nature—is close to zero within our accuracy. The dependence of the parameter $b(x) = [\alpha_{xy}(T) - \alpha_{xy}(0)]/T$ on these two quantities is of main interest. Figure 4(b) compares how $b(x)$ and n_h (determined [7] from ρ_{xy} above T_C) vary with x. Whereas, at small x, the decrease in $b(x)$ seems to match that of n_h, $b(x)$ falls much faster to zero at large x.

A striking relation between them becomes apparent if we plot one against the other. Figure 5 shows that, when x decreases below 0.85, $b(x)$ grows as a fractional power of $n_h - n_{h0}$ with n_{h0} a threshold density. This is consistent with $b(x)$ increasing as the density of states (DOS), viz., $b(x) \sim \mathcal{N}_F$. The DOS for the free-electron gas $\mathcal{N}_F^0$ (dashed curve) has a slightly stronger curvature than the data. Interestingly, the occurrence of the threshold doping at $x = 0.85$ accounts well for a puzzling change in the resistivity behavior when x exceeds 0.85 [Fig. 3(c)]. In general, slowly increasing x causes the resistivity profiles to change systematically, reflecting slight decreases in both n_h and ℓ_0 (mean-free path). However, between 0.85 and 1.0, the change is sudden and striking. At $x = 0.85$, ρ is T independent below 100 K consistent with a disordered metal. By contrast, at 1.0, ρ rises monotonically with decreasing T [Fig. 3(c)]. Between 300 and 4.2 K, ρ increases from 6.3 to 32 mΩ cm. At low T, conductivity proceeds by hopping between strongly localized states in an impurity band. Figure 5 confirms that we reach the extremum of the hole band near $x = 0.85$. Further removal of carriers ($x \to 1$) affects states within the impurity band.

Knowing n_h and ρ at each x, we may determine the mean-free path ℓ_0 in the impurity-scattering regime. Between $x = 0.1$ and 1.0, ℓ_0 decreases by a factor of 40. This steep decrease has no discernible influence on $b(x)$. Combining these factors then, we have $\alpha_{xy} = gT\mathcal{N}_F$, where g is independent of ℓ_0. We may boil down α_{xy} to the measurement of an "area" $\mathcal{A}$ by writing

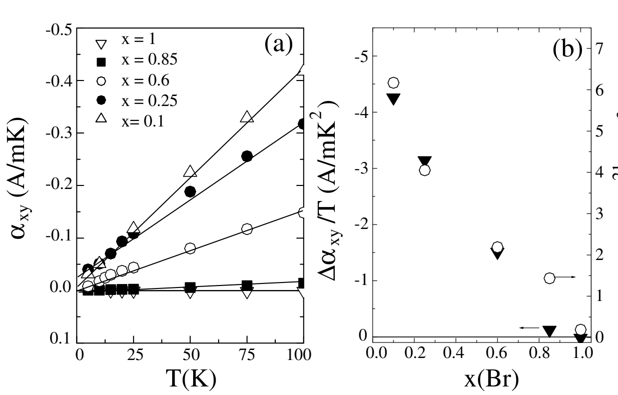

FIG. 4. (a) Curves of α_{xy} versus T obtained by adding e_N and $S\tan\theta_H$ [Eq. (1)]. The slope $b(x)$ now falls monotonically as x increases to 1.0. (b) compares how the slope $b(x) = \Delta\alpha_{xy}/T$ and n_h vary with x.

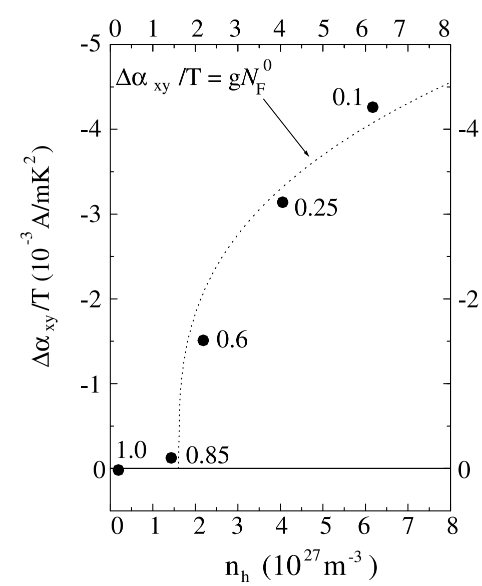

FIG. 5. Plot of $b(x) = \Delta\alpha_{xy}/T$ against n_h showing that for samples with $x \leq 0.85$ $b(x)$ increases as a fractional power of $n_h - n_{h0}$. The dashed line is $\Delta\alpha_{xy}/T = g\mathcal{N}_F^0$, with $g = 9.77 \times 10^{-50}$ in SI units.

$$\alpha_{xy} = \mathcal{A}\frac{ek_B^2 T}{\hbar}\mathcal{N}_F \qquad (T \ll T_C), \tag{4}$$

with k_B Boltzmann's constant and e the electron charge. The value of g gives $\mathcal{A} = 33.8$ Å^2 if we assume $\mathcal{N}_F \sim \mathcal{N}_F^0$. As the anomalous Hall heat current produced by $\mathbf{E}\|\hat{\mathbf{x}}$ is $J_y^Q = \alpha_{yx}TE$, it shares the simple form in Eq. (4). The ratio $J_y^Q/J_y \sim T^2$, as expected for a Fermi gas.

We briefly sketch the anomalous-velocity theory [2–4]. In a periodic lattice, the position operator for an electron is the sum $\mathbf{x} = \mathbf{R} + \mathbf{X}(\mathbf{k})$, where $\mathbf{R}$ locates a unit cell, while $\mathbf{X}(\mathbf{k})$ locates the intracell position [15]. A finite $\mathbf{X}(\mathbf{k})$ implies that $\mathbf{x}$ does not commute with itself. Instead, we have [15] $[x_j, x_k] = i\epsilon^{jkm}\Omega_m$, with ϵ^{jkm} the antisymmetric tensor, which implies the uncertainty relation $\Delta x_j \Delta x_k \sim \Omega$. The "'Berry curvature" $\mathbf{\Omega}(\mathbf{k}) = \nabla_{\mathbf{k}} \times \mathbf{X}$ is analogous to a magnetic field in $\mathbf{k}$ space [16]. In the presence of $\mathbf{E}$, $\mathbf{\Omega}$ adds a term that is the analog of the Lorentz force to the velocity $\mathbf{v}_{\mathbf{k}}$, viz.,

$$\hbar\mathbf{v}_{\mathbf{k}} = \nabla\epsilon(\mathbf{k}) - \mathbf{E} \times \mathbf{\Omega}(\mathbf{k}). \tag{5}$$

The anomalous-velocity term in Eq. (5) immediately implies the existence of a spontaneous Hall current $\mathbf{J}_H = -2e\sum_{\mathbf{k}} f_{\mathbf{k}}^0 \mathbf{E} \times \mathbf{\Omega}(\mathbf{k})$, where $f_{\mathbf{k}}^0$ is the unperturbed distribution [2–4,8,15]. The unconventional form of the current—notably the absence of any lifetime dependence—has made the KL theory controversial for decades [1,17]. However, strong support has been obtained from the measurements of Lee *et al.* [7] showing that the normalized AHE conductivity σ'_{xy}/n_h in $CuCr_2Se_{4-x}Br_x$ is unchanged despite a 1000-fold increase in ρ.

In general, the off-diagonal term α_{xy} is related to the derivative of σ_{xy} at the chemical potential μ, viz., $\alpha_{xy} = (\pi^2/3)(k_B^2 T/e)[\partial\sigma_{xy}/\partial\epsilon]_\mu$ [12]. Using the result [7] that σ'_{xy} is linear in n_h but independent of ℓ_0, and $[\partial n_h/\partial\epsilon]_\mu = \mathcal{N}_F$, we see that $\alpha_{xy} \sim T\mathcal{N}_F$, consistent with Eq. (4). (By contrast, we note that the skew-scattering model [17] would predict $\sigma'_{xy} \sim n_h\ell_0$ and $\alpha_{xy} \sim T\mathcal{N}_F\ell_0$.)

Finally, $\mathcal{A}$ in Eq. (4) has the value 34 Å^2. If Eq. (5) is indeed the origin of α_{xy}, $\mathcal{A}$ must be roughly the scale of $\Omega \sim \Delta x_j \Delta x_k$. Hence the value $\mathcal{A} \sim \frac{1}{3}\times$ the unit-cell area seems reasonable (the lattice spacing here is 10.33 Å). While a quantitative comparison requires knowledge of $\mathbf{\Omega}(\mathbf{k})$ over the Brillouin zone, the simple form of Eq. (4) seems to provide valuable insight on the anomalous heat current.

A previous calculation of the Nernst coefficient was based on the "side-jump" model [18]. On scattering from an impurity, the carrier suffers a small sideways displacement δ to give on average $\tan\theta_H = \delta/\ell_0$. This was used to derive $Q_s \sim T(k_F\ell)^{-1}$. In our experiment, $k_F\ell$ falls monotonically, with increasing x, while e_N rises to a broad maximum near 0.25 before falling. Hence our experiment is in essential conflict with the side-jump model. From earlier experiments [11], an empirical form $Q_s = -(a + b'\rho)T$ has been inferred (a, b' are constants). This is not borne out in our data.

Combining Nernst, Hall, and thermopower experiments on the ferromagnet $CuCr_2Se_{4-x}Br_x$, we have determined how α_{xy} (hence $\mathbf{J}^Q$) changes as a function of n_h and τ. At low T, we find that α_{xy} follows the strikingly simple form $\alpha_{xy} \sim \mathcal{A}T\mathcal{N}_F$, consistent with the anomalous-velocity theory for the AHE (Fig. 4). In addition, a direct relation [Eq. (3)] between M and α_{xy} is observed in the paramagnetic regime above T_C.

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