Anomalous Hall Heat Current and Nernst Effect in the CuCr$_2$Se$_4$-$x$Br$_x$ Ferromagnet

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In a ferromagnet, an anomalous Hall heat current, given by the off-diagonal Peltier term $\alpha_{xy}$, accompanies the anomalous Hall current. By combining Nernst, thermopower, and Hall experiments, we have measured how $\alpha_{xy}$ varies with hole density and lifetime $\tau$ in CuCr$_2$Se$_4$-$x$Br$_x$. At low temperatures $T$, we find that $\alpha_{xy}$ is independent of $\tau$, consistent with anomalous-velocity theories. Its magnitude is fixed by a microscopic geometric area $\mathcal{A}$, $\alpha_{xy} \sim 34 \AA^2$. Our results are incompatible with some models of the Nernst effect in ferromagnets.

In a ferromagnet, the anomalous Hall effect (AHE) is the appearance of a spontaneous Hall current flowing parallel to $E \times M$, where $E$ is the electric field and $M$ 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$_2$Se$_4$-$x$Br$_x$. Lee et al. [7] reported that, despite a 1000-fold increase in the resistivity $\rho$ induced by varying the Br content, 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 $J^0$ also accompanies the AHE current in the absence of any temperature gradient [9,10]. In principle, $J^0$ 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 $E_N$ parallel to $M \times (-\nabla T)$. However, in previous Nernst experiments on ferromagnets [9–11], $J^0$ was not found because other transport quantities were not measured. Combining the Nernst signal with the AHE resistivity $\rho_{yx}$ and the thermopower, we have determined how the transport quantity $\alpha_{xy}$ relevant to $J^0$ varies in CuCr$_2$Se$_4$-$x$Br$_x$ as the hole density $n$ and carrier lifetime $\tau$ are greatly changed under doping. We show that $\alpha_{xy}$ has a strikingly simple form, with its magnitude scaled by a microscopic geometric area $\mathcal{A}$.

We apply a gradient $-\nabla T$ on an electrically isolated sample in a magnetic field $H$ [2]. Along $\hat{x}$, 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 $\hat{y}$, however, both $E_x$ and $-\nabla T$ generate Hall-type currents. In general, the charge current in the presence of $E$ and $-\nabla T$ is $\mathbf{J} = \mathbf{E} + \mathbf{J}_x + \mathbf{J}_y$, with $\mathbf{E}$ and $\mathbf{J}_x$ the electrical and thermoelectric (“Peltier”) conductivity tensors, respectively. Setting $\mathbf{J}_y = 0$, we obtain the Nernst signal $e_N = E_y/|\nabla T| = \rho \alpha_{xy} + \rho_{yx} \alpha_x$, where $\alpha = \alpha_{xy}$ [12]. Hence, as noted, the Nernst signal results from the two distinct $y$-axis charge currents, $\alpha_{yx} (-\nabla T)$ and $\alpha_{xy} E_x$.

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 $\alpha_{xy}$ as

$$\rho \alpha_{xy} = e_N + S \tan \theta_H. \quad (1)$$

![FIG. 1. Curves of the measured $e_N = E_x/|\nabla T|$ versus $H$ in CuCr$_2$Se$_4$-$x$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 \( \alpha_{xy} \), we need to measure \( e_N, S, \rho_{xy}, \) and \( \rho \). Knowing \( \alpha_{xy} \), we readily find the transverse heat current
\[
J_T^Q = \tilde{\alpha}_{xy} E_x,
\]
since \( \tilde{\alpha}_{xy} = \alpha_{xy} T \) by Onsager reciprocity.

The spinel CuCr$_2$Se$_4$ 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 \( n_h \) increases from 0 to 1, the value of \( \rho_{xy} \) at 5 K increases by \( \sim 10^3 \), while \( \rho_{xy}'/\rho_{xy} \) 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 \( \rho, M, \) and \( \rho_{xy}' \) 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

\[
e_N = Q_0 \mu_0 H + Q_s \mu_0 M.
\]

For \( T < T_C \) in all samples, the \( Q_0 \) term cannot be resolved, so that \( e_N \approx Q_s \mu_0 M \). 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

\[
\alpha_{xy} = \beta M \quad (T > T_C),
\]

where \( \beta \) is only weakly \( T \) dependent (it decreases by 5% between 250 and 400 K). The parameter \( \beta \) plays the important role of relating the magnitudes of the paramagnetic \( M \) and the transverse electronic current (through the Nernst signal). Its minuscule value \( (\beta \approx 2 \times 10^{-7} \text{ K}^{-1} \) 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).](image)

![FIG. 3. (a) Curves of \( e_N \) versus \( T \) below 150 K in five samples with doping 0.1 \( \leq x \leq 1.0 \) showing nominal \( T \)-linear behavior at low \( T \) (\( H = 2T \)). 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 \( \rho-T \) profiles in the samples with \( x = 0.85 \) and 1.0 (\( H = 0.0 \)). At low \( T, \rho \) at 0.85 is metallic, but at 1.0 \( \rho \) reveals hopping between strongly localized states.](image)
increases to 1.0. (b) compares how the slope $b$ versus $x$ increases from 0.1 to 1.0 (Fig. 4). In all samples except $x = 0.1$ and 1.0, the change is sudden and striking. At $x = 0.85$, $\rho$ is $T$ independent below 100 K consistent with a disordered metal. By contrast, at 1.0, $\rho$ rises monotonically with decreasing $T$ [Fig. 3(c)]. Between 300 and 4.2 K, $\rho$ increases from 6.3 to 32 m$\Omega$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 ($x \rightarrow 1$) affects states within the impurity band.

Knowing $n_h$ and $\rho$ at each $x$, we may determine the mean-free path $\ell_0$ in the impurity-scattering regime. Between $x = 0.1$ and 1.0, $\ell_0$ 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 $\ell_0$. We may boil down $\alpha_{xy}$ to the measurement of an “area” $\mathcal{A}$ by writing $\Delta\alpha_{xy}/T = g\mathcal{N}_F^0$.
\[ \alpha_{xy} = A \frac{e^{k_{B}T}}{h} N_F \quad (T \ll T_C), \tag{4} \]

with \( k_B \) Boltzmann’s constant and \( e \) the electron charge. The value of \( g \) gives \( A = 33.8 \text{ Å}^2 \) if we assume \( N_F \sim N_F \). As the anomalous Hall heat current produced by \( E | \mathbf{k} \rangle = \alpha_{xy} E \mathbf{F} \), it shares the simple form in Eq. (4). The ratio \( J_y^0/J_y \sim T^2 \), 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 \[15\] \[ x_j, x_k \] = \( i e^{i k m} \Omega_{mn} \), with \( e^{i k m} \) the antisymmetric tensor, which implies the uncertainty relation \( \Delta x_j \Delta x_k \sim \Omega \). The “Curry curvature” \( \Omega(\mathbf{k}) = \nabla_R \times \mathbf{X} \) is analogous to a magnetic field in space \[16\]. In the presence of \( E, \Omega \) adds a term that is the analog of the Lorentz force to the velocity \( v_k \), viz.

\[ h v_k = \mathbf{E}(\mathbf{k}) - E \times \mathbf{\Omega}(\mathbf{k}). \tag{5} \]

The anomalous-velocity term in Eq. (5) immediately implies the existence of a spontaneous Hall current \( J_H = -2e \sum_k \Re E \times \mathbf{\Omega}(k) \), where \( J_H \) 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 normal AHE conductivity \( \sigma_{xy}/n_h \) in CuCr\(_2\)Se\(_4\)–Br\(_x\) is unchanged despite a 1000-fold increase in \( p \).

In general, the off-diagonal term \( \alpha_{xy} \) is related to the derivative of \( \sigma_{xy} \) at the chemical potential \( \mu \), viz. \( \alpha_{xy} = (\pi^2/3)(k_B^2 T/e) [\partial \sigma_{xy}/\partial \mu]_\mu \) \[12\]. Using the result \[7\] that \( \sigma_{xy} \) is linear in \( n_h \) but independent of \( \ell_0 \), and \( \partial n_h/\partial \mu = N_F \), we see that \( \alpha_{xy} \sim T \partial N_F \), consistent with Eq. (4). (By contrast, we note that the skew-scattering model \[17\] would predict \( \sigma_{xy} \sim n_h \ell_0 \) and \( \alpha_{xy} \sim T \partial N_F \ell_0 \).

Finally, \( A \) in Eq. (4) has the value 34 Å\(^2\). If Eq. (5) is indeed the origin of \( \alpha_{xy} \), \( A \) must be roughly the scale of \( \Omega \sim \Delta x_j \Delta x_k \). Hence the value \( A \sim 1 \times \) 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 \( \delta \) to give on average \( \tan \mu \gamma = \delta / \ell_0 \). This was used to derive \( Q_y = (k_r/F \ell)^{-1} \). In our experiment, \( k_r \ell \) 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 \( Q_y = -(a + b') T \) 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\(_2\)Se\(_4\)–Br\(_x\), we have determined how \( \alpha_{xy} \) changes as a function of \( n_h \) and \( T \). At low \( T \), we find that \( \alpha_{xy} \) follows the strikingly simple form \( \alpha_{xy} \sim AT N_F \), consistent with the anomalous-velocity theory for the AHE (Fig. 4). In addition, a direct relation [Eq. (3)] between \( M \) and \( \alpha_{xy} \) is observed in the paramagnetic regime above \( T_C \).

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