Anomalous Hall effect due to magnetic chirality in the Pyrochlore lattice

A. Kalitsov, B. Canals and C. Lacroix

Institut Néel, CNRS-UJF, 25 avenue des Martyrs, bât. D, BP 166, 38042 Grenoble Cedex 9, France

E-mail: alan.kalitsov@grenoble.cnrs.fr

Abstract. We calculate the intrinsic Hall conductivity of conduction electrons coupled to a pyrochlore lattice of localized moments. In the presence of anisotropy the localized moments may order in a non-coplanar chiral structure, which can be at the origin of an intrinsic Hall effect. The sign and the value of the Hall conductance depend on the band filling factor and the local exchange coupling. Application of an external magnetic field induces a change in the magnetic structure of the localized spins which results in a change of the Hall conductance. In this framework, the temperature dependence of the Hall conductance may exhibit a nonmonotonical behavior. The results are in a qualitative agreement with experimental results on Mo pyrochlores.

1. Introduction

The anomalous Hall effect (AHE) in ferromagnetic systems has attracted great interest both in the field of fundamental science and practical applications. The origin of AHE is spin-orbit interaction in metallic ferromagnets [1, 2, 3]. Recently an unusual behavior of AHE has been found experimentally in manganites [4, 5], pyrochlore molybdates [6, 7, 8, 9, 10, 11] and iridates [12, 13, 14].

The pyrochlore \( A_2B_2O_7 \) lattice is composed of two sublattices of atom \( A \) and atom \( B \), for example molybdates \( \text{Nd}_2\text{Mo}_2\text{O}_7 \), \( \text{Gd}_2\text{Mo}_2\text{O}_7 \), or the iridate \( \text{Pr}_2\text{Ir}_2\text{O}_7 \). The sublattices are structurally identical but are displaced by half a lattice constant from each other. The ions \( A \) and \( B \) locate at the vertex of corner-sharing tetrahedra in each sublattice. The experiments show that the AHE conductivity in pyrochlores changes sign with increasing the magnetic field applied along [111] direction [6, 7]. The temperature dependence of AHE conductance indicates that it may vary from continuous increase on cooling in \( \text{Nd}_2\text{Mo}_2\text{O}_7 \) [6, 7] or \( \text{Pr}_2\text{Ir}_2\text{O}_7 \) [12] to nonmonotonical behavior for \( \text{Gd}_2\text{Mo}_2\text{O}_7 \) when its absolute value is maximum at about half of the Curie temperature [7].

The AHE behavior in these systems cannot be understood in the framework of the earlier theories [1, 2, 3]. A new intrinsic mechanism, related to the noncoplanar spin configuration has been proposed [15, 16, 17, 18, 19]. This mechanism is related to the spin chirality of the conduction electron spins coupled to the localized spins at each site of the lattice with a local exchange coupling \( J_0 \).

Ohgushi et al. [15] calculated the AHE for a two-dimensional electron gas moving on a Kagomé lattice in the limit of \( J_0 \to \infty \). Taillefumier et al. [16] calculated the AHE in a Kagomé
lattice with finite exchange coupling and showed that the value of $J_0$ can distinguish between two different regimes associated to different Chern numbers.

In real systems electrons are moving in three-dimensions. The AHE theory was generalized for three-dimensions by Montambaux [20], Goryo and Kohmoto [21] as well as by Onoda and Nagaosa [18]. Onoda and Nagaosa calculated the AHE in the presence of the fluctuating spin chirality of the localized spins. They show that the spin chirality mechanism works even without the correlations of the localized spins [18].

The behavior of AHE in three-dimensions can be very different from that in two-dimensions because of the difference in the band structure. The aim of this work is to study the AHE in a realistic model closer to the experimental situation of pyrochlores and to infer whether the spin chirality mechanism on its own may account for the observed anomalies of the Hall conductance.

2. Model
2.1. Localized moments on a pyrochlore lattice: magnetic structure in presence of local anisotropy and external magnetic field
We consider a model of conduction electrons coupled to classical localized moments $\mathbf{S}_i$ with strong on-site uniaxial anisotropy and ferromagnetic exchange on a pyrochlore lattice. Due to the anisotropy and ferromagnetic exchange, the localized spins system behaves as a spin ice and from now on, we assume that this spin-ice system orders in a $q = 0$ structure (as Nd$_2$Mo$_2$O$_7$ does [6, 7]). In the absence of external field $\mathbf{B}$, the spins $\mathbf{S}_i$ on each tetrahedron are supposed to form a "two in, two out" ordered magnetic structure (see Fig. 1 a). If magnetic field is applied along the [111] direction, this structure changes to the "one in, three out" configuration above some critical field (Fig. 1 b), where we still assume a $q = 0$ ordering.

The Hamiltonian of the localized moments is written as:

$$H_{loc} = -I_0 \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j - D_0 \sum_i (\mathbf{n}_i \cdot \mathbf{S}_i)^2 - g\mu_B \mathbf{B} \cdot \sum_i \mathbf{S}_i,$$

(1)

where $I_0$ is the ferromagnetic exchange coupling between localized spins, $D_0$ is the uniaxial anisotropy constant, $\mathbf{n}_i$ is a unit vector directed along the local [111] axis and $\mathbf{B}$ is the magnetic field applied along the [111] direction. The spins $\mathbf{S}_i$ are considered here as classical spins and the magnetic structure is determined by minimizing the energy given by Eq.(1). There are three energetically equivalent spin configurations "in - in - out - out", "in - out - in - out" and "in - out - out - in" when an infinitesimal magnetic field is applied along [111] direction. We choose "in - in - out - out" as the initial spin configuration. On Fig. 2 we plot the components of the localized spin $\mathbf{S}_2$ as a function of the external field. $\mathbf{S}_2$ flips at the critical field $\| \mathbf{B}_c \| = I_0/\sqrt{3}g\mu_B$ and the spin structure becomes "in - out - out - out" (Fig. 1 b).

2.2. Anomalous Hall Effect
We consider conduction electrons coupled to the localized spins and described in the tight binding approximation. The Hamiltonian of these conduction electrons writes

$$H = t \sum_{i,j,\sigma} (c_i^{\sigma\dagger} c_j^{\sigma} + h.c.) - \sum_i c_i^{\uparrow \dagger} (\mathbf{\tau}_{\alpha\beta} \cdot (J_0 \mathbf{S}_i + g\mu_B \mathbf{B})) c_i^{\beta},$$

(2)

where $t$ is the hopping integral between two neighboring sites, $c_i^{\sigma}$ and $c_i^{\sigma\dagger}$ are the creation and annihilation operators of the conduction electron with spin $\sigma$ on site $i$, $\mathbf{\tau}_{\alpha\beta}$ describes the Pauli matrices, and $J_0$ is the local exchange between conduction electrons and localized spins.

Making the Fourier transform, we rewrite the Hamiltonian (2) in the reciprocal space, $H(\mathbf{k})$, and use the Kubo formula [16] to calculate the anomalous Hall conductance

$$\sigma_{\alpha\beta}(\omega) = \frac{e^2 \hbar}{V} \sum_{n, \mathbf{k}} \sum_{m \neq n} \frac{(v_{n})^{mn}(v_{\beta})^{mn} - (v_{\alpha})^{mn}(v_{\beta})^{mn}}{\epsilon_{n, \mathbf{k}} - \epsilon_{m, \mathbf{k}}},$$

(3)
where $V$ is the volume of reciprocal lattice unit cell, $\epsilon_{n,k}$ is the eigenvalue of the Hamiltonian $H(k)$ corresponding to the energy of the $n$th band for the wave vector $k$, $f_{n,k}$ is the Fermi-Dirac distribution function and $(v_\alpha)^{nm} = \hbar^{-1} < n,k | (\partial H(k)/\partial k_\alpha) | m,k >$ is the matrix element of the $\alpha$ component of the electrons velocity operator ($\alpha, \beta = x, y, z$).

We introduce the temperature dependence of AHE by assuming that the magnitude of each local moment follows a mean field-type variation $S_i(T) = S_i^{(0)} (1 - T/T_c)^{1/2}$, where $\|S_i^{(0)}\| = 1$, $T_c$ is the critical temperature of the associated second order phase transition. It therefore replaces in Eq. (1) and (2) the zero-temperature exchange and anisotropy constants $I_0$, $J_0$ and $D_0$ by temperature-dependent exchange and anisotropy constants $I(T) = I_0 (1 - T/T_c)$, $J(T) = J_0 (1 - T/T_c)^{1/2}$ and $D(T) = D_0 (1 - T/T_c)^{3/2}$ as well as renormalizing magnetic field vector in the Hamiltonian (1) $B = B_0 (1 - T/T_c)^{1/2}$. The critical field also becomes temperature dependent.

3. Numerical results and discussion

In the Kagomé lattice it was found [15, 16] that there exists a range of parameters for which the 6 bands are separated by gaps, and it was shown that this may lead to quantized Hall effect. In the pyrochlore, this is no longer the case. There are eight bands which become splitted into two groups of four bands for a finite $J_0$ but each group does not have band gaps even for large values of $J_0$, therefore preventing any quantization of conductivity.

In three dimensions the AHE conductance has three components $\sigma_{xy}$, $\sigma_{xz}$ and $\sigma_{yz}$. With the choice of the "in - in - out - out" as the initial spin configuration, we obtain $\sigma_{xz} = 0$ and $\sigma_{yz} = 0$ at zero magnetic field. If the magnetic field applied along the [111] direction all components of the AHE conductance become finite but $\sigma_{yz}$ remain equivalent.

The calculations were done with the following model parameters. We set $t = 0.25 eV$ which corresponds to half of the bandwidth $W = 8t = 2 eV$ calculated for Gd$_2$Mo$_2$O$_7$ [22]. The other parameters were taken as: $J_0 = 2t$, $g = 1.71$ [10], $I_0 = 2 \cdot 10^{-4} eV$ which roughly corresponds to the experimental value of the Curie temperature $T_c = 90 K$ [7, 8] for Nd$_2$Mo$_2$O$_7$ compounds.

First we study the field dependence of the AHE conductance for different values of the band
filling factor \( p \) corresponding to the number of conduction electrons per tetrahedron. On Fig. 3 and Fig. 4 we plot the field dependence of the AHE conductance for \( p = 1/8 \) and \( p = 7/8 \) respectively. The value of the AHE conductance for a large anisotropy constant \( D_0 \) behaves as a step-function and changes sign at the critical field which corresponds to the transition from "two-in, two-out" to "one-in, three-out" spin structure. When the spin structure becomes "one-in, three-out" the three components of the conductivity are equivalent and therefore \( \sigma_{xy} = \sigma_{xz} = \sigma_{yz} \).

**Figure 3.** Field dependence of AHE conductance with \( p = 1/8, D_0 = 10 I_0, T = 0 \text{K} \).

**Figure 4.** The same as in Fig. 3 with \( p = 7/8 \).

We found that the behavior of the field dependence of AHE conductance for smaller value of the anisotropy constant is similar. The small difference in the slopes of \( \sigma_{\alpha\beta} \) is coming from the different slopes of \( S_i(B) \), which are themselves implicit function of the on site anisotropy. Comparing the results shown on Fig. 3 and Fig. 4 one can conclude that AHE conductance strongly depends on the band filling factor. Change from almost empty bands (\( p = 1/8 \), Fig. 3) to almost filled bands (\( p = 7/8 \), Fig. 4) leads to a change of the magnitude and the sign of AHE conductance. In case of half-filling (\( p = 1/2 \)) all components of AHE conductance becomes zero because the Fermi energy lies in the gap between the two groups of four bands.

In order to discuss the relation between the spin chirality and AHE conductance, we define the fictitious magnetic flux as the sum of four vectors \( \Phi_i \) whose directions are along the normal vectors \( \mathbf{n}_{jkl} \), perpendicular to the triangle formed by the sites of three spins \( j, k, \) and \( l \) [6, 12]

\[
\Phi = \sum_i \Phi_i = \sum_i \chi_i \mathbf{n}_{jkl},
\]

where the scalar chirality \( \chi_i \) is defined for each triangle as

\[
\chi_i = S_j \cdot (S_k \times S_l),
\]

where \( j, k, l \) is the \( i \)th set of different numbers chosen in a cyclic way from 1-4 to specify the set of spins within a tetrahedron. We find that for some set of the model parameters, the Hall conductance is proportional to the fictitious magnetic flux. However this relation does not generally hold. A similar relation between the Hall conductance and the spin chirality has been shown by Tatara and Kawamura in the weak coupling limit [17] as well as in the intermediate and in the strong coupling limits in the antiferromagnetic Kagomé lattice [16].
On Fig. 5 and Fig. 6 we plot the $J_0$ dependence of AHE conductance at zero temperature and zero magnetic field that yields $\sigma_{xz} = 0$ and $\sigma_{yz} = 0$. $\sigma_{xy}$ is the only non-zero component of AHE conductance. When the band filling factor $p = 1/8$ (Fig. 5) $\sigma_{xy}$ monotonically increases with $J_0$ and reaches a plateau at $J_0 = 0.8$ eV. On contrast, the $J_0$ dependence of AHE conductance for $p = 5/8$ (Fig. 6) is nonmonotonical. It has a maximum at $J_0 = 0.2$ eV, changes its sign at $J_0 = 0.3$ eV and it varies smoothly when $J_0 > 0.4$ eV. Increase of a local exchange coupling $J_0$ corresponds to decrease of temperature.

![Figure 5](image1.png)

**Figure 5.** $J_0$ dependence of AHE conductance with $D_0 = 10I_0$, $T = 0$ K, $B = 0$ T and $p = 1/8$.

On Fig. 7 and Fig. 8 we show the temperature dependence of the AHE conductance for $p = 1/8$ and $p = 5/8$ respectively. In our model the increase of temperature lead to two effects, first the transition from "two – in, two – out" to "one – in, three – out" spin structure at about $T = 60$ K, second decreasing of the exchange coupling $J(T)$. Comparing the results shown on Fig. 7 and Fig. 8 one can conclude that the temperature dependence of the AHE conductance strongly depends on the band filling factor and it exhibits a nonmonotonal behavior which corresponds either to the transition from "two – in, two – out" to "one – in, three – out" spin structure at about $T = 60$ K (Fig. 7) or decrease of the exchange coupling $J(T)$ at about $T = 70$ K (Fig. 8).

Concerning Nd$_2$Mo$_2$O$_7$, experiments involving neutron scattering on single crystals, as a function of field and temperature, coupled to thermodynamics probes like magnetization and specific heat, led to the conclusion that the spin chirality mechanism cannot be the only one to account for the anomalous transport observed in this compound.

The results obtained in this work cannot fully address this system because in real materials, AHE originates from both intrinsic and extrinsic mechanisms. The extrinsic mechanism is the asymmetric scattering of spin-polarized electrons on impurities due to spin-orbit interactions. It is the intrinsic mechanism due to non-coplanar spin configurations that was considered in this paper. However this mechanism is a relevant mechanism of AHE in pyrochlore compounds because the calculated value of the AHE conductance in the units of $\Omega^{-1} cm^{-1}$ is comparable with the experimental value $[6, 7, 12]$.

Within our framework, we have shown that AHE conductance is strongly connected to the non coplanar spin structure but is not necessarily proportional to the spin chirality.

These results are in qualitative agreement with experimental results on the molydbdates pyrochlore Nd$_2$Mo$_2$O$_7$. 

![Figure 6](image2.png)

**Figure 6.** The same as in Fig. 5 with $p = 5/8$. 

Highly Frustrated Magnetism 2008 (HFM 2008) IOP Publishing 
Journal of Physics: Conference Series 145 (2009) 012020 doi:10.1088/1742-6596/145/1/012020
Figure 7. Temperature dependence of AHE conductance with $D_0 = 4I_0$, $B = \sqrt{3} \cdot 0.6T$ and $p = 1/8$.

Figure 8. The same as in Fig.7 with $p = 5/8$.

4. References

[1] Karplus R and Luttinger J M 1954 Phys. Rev. 95 1154
[2] Smit J 1955 Physica (Amsterdam) 21 877
[3] Berger L 1970 Phys. Rev. B 2 1459
[4] Matl P, Ong N P, Yan Y F, Li Y Q, Studebaker D, Baum T and Doubinina G 1998 Phys. Rev. B 57 10248
[5] Ye J, Kim Y B, Milles A J, Shraiman B I, Majumdar R and Tesanovic Z 1999 Phys. Rev. Lett. 83 3737
[6] Taguchi Y, Oohara Y, Yoshizawa H, Nagaosa N and Tokura Y 2001 Science 291 2573
[7] Taguchi Y, Sasaki T, Awaji S, Iwasa Y, Tayama T, Sakakibara T, Iguchi S, Ito T and Tokura Y 2003 Phys. Rev. Lett. 90 257202
[8] Yoshii S, Ikubo S, Kageyama T, Oda K, Kondo Y, Murata K and Sato M 2000 J. Phys. Soc. Jpn. 69 3777
[9] Yasui Y, Kondo Y, Kanada M, Ito M, Harashina H, Sato M and Kakurai K 2001 J. Phys. Soc. Jpn. 70 284
[10] Yasui Y, Ikubo S, Harashina H, Kageyama T, Ito M, Sato M and Kakurai K 2003 J. Phys. Soc. Jpn. 72 865
[11] Yasui Y, Kageyama T, Moyoshi T, Soda M, Sato M and Kakurai K 2006 J. Magn. Magn. Mat. 310 e544
[12] Machida Y, Nakatsuji S, Maeno Y, Tayama T, Sakakibara T and Onoda S 2007 Phys. Rev. Lett. 98 057203
[13] Nakatsuji S, Machida Y, Maeno Y, Tayama T, Kakurai K, van Duijn J, Balicas L, Millican J N, Macalusa R T and Cham J Y 2006 Phys. Rev. Lett. 96 087204
[14] Yanagishima D and Maeno Y 2001 J. Phys. Soc. Jpn. 70 2880
[15] Ohgushi K, Murakami S and Nagaosa N 2000 Phys. Rev. B 62 R6065
[16] Taillefumier M, Canals B, Lacroix C, Dugaev V K and Bruno P 2006 Phys. Rev. B 74 085105
[17] Tatara G and Kawamura H 2002 J. Phys. Soc. Jpn. 71 2613
[18] Onoda S and Nagaosa N 2003 Phys. Rev. Lett. 90 196602
[19] Nagaosa N 2003 J. Phys. Soc. Jpn. 72 042001
[20] Montambaux G and Kohmoto M 1990 Phys. Rev. 41 11417
[21] Goryo J and Kohmoto M 2002 J. Phys. Soc. Jpn. 71 1403
[22] Solovyev I V 2003 Phys. Rev. B 67 174406