# Anomalous Hall Effect as a Probe of the Chiral Order in Spin Glasses

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The anomalous Hall effect arising from the noncoplanar spin configuration (chirality) is discussed as a probe of the chiral order in spin glasses. It is shown that the Hall coefficient yields direct information about the linear and nonlinear chiral susceptibilities of the spin sector, which has been hard to obtain experimentally from the standard magnetic measurements. Based on the chirality scenario of spin-glass transition, predictions are given on the behavior of the Hall resistivity of canonical spin glasses.

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For decades, spin glasses have been extensively studied as a prototype of "complex" systems characterized by both "frustration" and "randomness" [1]. Among a wide variety of spin-glass (SG) materials, most familiar and well studied is perhaps the so-called canonical SG, a dilute noble metal/$3d$ transition metal alloys. In canonical SG, the interaction between localized moments is the RKKY interaction which is mediated by conduction electrons via the $s$-$d$ exchange coupling $J_{sd}$. The oscillating nature of the RKKY interaction with distance, combined with spatially random arrangement of localized moments, gives rise to frustration and randomness. Since the RKKY interaction is isotropic in spin space, canonical SG, similar to many other SGs, is nearly isotropic in spin space, and is expected to be well modeled by the Heisenberg model. Weak magnetic anisotropy is mostly due to the Dzyaloshinski-Moriya (DM) interaction caused by the combined effect of the $s$-$d$ coupling and the spin-orbit interaction. The nearly isotropic character of the magnetic interaction in canonical SG is in apparent contrast to most of theoretical approaches which have been based on the Ising model describing the extremely anisotropic limit [1].

Experimentally, it is now well established that typical SG magnets including canonical SG exhibit an equilibrium phase transition at a finite temperature and there exists a thermodynamic SG phase. The true nature of the SG transition and of the SG ordered state, however, still remains elusive in spite of extensive studies [1].

Although standard theories of the SG order invoke the Ising model as a minimal model, a scenario very different from the standard picture was proposed by the present author, which may be called a chirality scenario [2,3]. In this scenario, *chirality*, which is a multispin quantity representing the sense or the handedness of local noncoplanar structure of Heisenberg spins, plays an essential role. The local chirality may be defined for three neighboring Heisenberg spins by the scalar,

$$\chi_{ijk} = \vec{S}_i \cdot \vec{S}_j \times \vec{S}_j. \tag{1}$$

The chirality defined above is often called a scalar chirality: It takes a nonzero value when the three spins span the noncoplanar configuration in spin space, whose sign is representing the handedness of such noncoplanar spin configuration.

The chirality scenario of SG transition consists of two parts [2,3]: In a fully isotropic Heisenberg SG, it claims the occurrence of a novel *chiral-glass* ordered state in which only the chirality exhibits a glassy long-range order keeping the Heisenberg spin paramagnetic (spin-chirality decoupling). In a weakly anisotropic Heisenberg SG, the scenario claims that the weak random magnetic anisotropy "recouples" the spin to the chirality, and the chiral-glass order of the fully isotropic system shows up in the spin sector as the standard SG order via the magnetic anisotropy (spin-chirality recoupling). In other words, the experimental SG transition and the SG ordered state are the chiral-glass transition and the chiral-glass ordered state of the fully isotropic system "revealed" by the weak random magnetic anisotropy inherent to real SG.

Thus far, an experimental test of the chirality scenario remains indirect. This is mainly due to the experimental difficulty in directly measuring the chirality. A possible clue to overcome this difficulty was recently found via the study of electron transport properties of certain magnets in which conduction electrons interact with the core spins possessing chiral degrees of freedom. In particular, it has been realized that under appropriate conditions a chirality contribution shows up in the anomalous part of the Hall effect. This was first pointed out in the strong coupling case where the conduction electrons are strongly coupled with core spins via the Hund coupling, as in the cases of manganites [4,5] and in frustrated Kagomé [6], or pyrochlore ferromagnet [7,8] (see also Ref. [9]). In the weak-coupling case which is more relevant to canonical SG, chirality contribution to the anomalous Hall effect was examined by Tatara and Kawamura [10]. By applying the linear response theory and the perturbation expansion to the standard $s$-$d$ Hamiltonian, these authors derived the chirality contribution to the Hall resistivity.

Under such circumstances, the purpose of the present Letter is twofold. First, on the basis of the formula derived in Ref. [10], I wish to explore in some detail the properties of the Hall resistivity at the SG transition with particular interest in the chirality contribution, and propose the way to extract information about the chiral order of the spin sector from the experimental data. Second, on the basis of the aforementioned chirality theory of SG transition, I present several predictions on the expected behavior of the anomalous part of the Hall resistivity, which might serve as an experimental test of the chirality theory. In the following, I will discuss these two issues successively.

Conduction electrons on the lattice with $N$ sites are coupled with core spins (assumed to be classical and fixed) via the standard $s$-$d$ exchange interaction $J_{sd}$, and are also scattered by normal impurities. Assuming the weak-coupling regime in which $J_{sd}$ is smaller than the Fermi energy $\epsilon_F$, the first nonzero contribution to the Hall conductivity comes from the third-order term in the perturbation, which can be recast into the Hall resistivity as

$$\rho_{xy}^{(\mathrm{chiral})} = 54\pi^2 \chi_0 \left(\frac{J_{sd}}{\epsilon_F}\right)^2 J_{sd}\tau\rho_0 = CJ_{sd}^3\chi_0, \tag{2}$$

$$\chi_0 = \frac{1}{6Nk_F^2}\sum_{ijk}\chi_{ijk}\left[\frac{(\vec{r}_{ij}\times\vec{r}_{jk})_z}{r_{ij}r_{jk}} I'(r_{ij})I'(r_{jk})I(r_{ki}) + (\text{two permutations})\right], \tag{3}$$

where $\rho_0$ is the Boltzmann resistivity, $\tau$ is the mean collision time, $k_F$ is the Fermi wave number, and $\chi_{ijk}$ represents the local chirality defined by Eq. (1), $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$, etc., with $r_{ij} \equiv |\vec{r}_{ij}|$, etc. $I(r)$ represents a function decaying as $I(r) = [(\sin k_F r)/(k_F r)]e^{-r/2\ell}$, with $\ell$ being the electron mean-free path, and $I'(r) = [dI(r)]/(dr)$. One sees from Eq. (3) that $\chi_0$ is a total (net) chirality, while the factor in the square bracket in Eq. (3) specifies the coupling between the spin space and the real space. In canonical SG, $J_{sd}$ is positive. The coefficient $C$ is positive in the single-band approximation but, more generally, its sign would depend on the detailed band structure of the material.

By contrast, conventional theories of the anomalous Hall effect have attributed its origin to the spin-orbit interaction $\lambda$ and a finite magnetization $M$ [11–13], i.e., mechanisms known as the skew scattering or the side jump. Note that the chirality contribution is independent of these conventional ones. Taking account of the conventional terms within the perturbation scheme, the anomalous part of the Hall resistivity has been given by

$$\begin{aligned}\rho_{xy} &= -\lambda M(A\rho + B\rho^2) + CJ_{sd}^3\chi_0 \\ &= -M(\tilde{A}\rho + \tilde{B}\rho^2) + \tilde{C}\chi_0,\end{aligned} \tag{4}$$

where $\rho$ is the longitudinal resistivity $\rho = \rho_{xx}$, $A$ and $B$ are constants both positive within the single-band approximation [10], and $\tilde{A} = A\lambda$, $\tilde{B} = B\lambda$, and $\tilde{C} = CJ^3$.

Since Heisenberg spins are frozen in a spatially random manner in the SG ordered state, the sign of the local chirality appears randomly, which inevitably leads to the vanishing total chirality in the bulk, $\chi_0 = 0$. It thus appears that the chirality-driven anomalous Hall effect vanishes in bulk SG samples. In the strong coupling case, however, a mechanism out of this cancellation was proposed by Ye *et al.* [4]. These authors pointed out that the spin-orbit interaction $\lambda$ in the presence of a net magnetization $M$ contains a term of the form

$$\mathcal{H}_{so} \approx \tilde{D}M\chi_0, \tag{5}$$

which, in the spin Hamiltonian, couples the total chirality to the total magnetization. In the weak-coupling regime relevant to canonical SG, a term of the form (5) with $\tilde{D} = D\lambda(J_{sd}/\epsilon_F)^2(J\tau)^2$ was also derived perturbatively by taking the electron trace of the spin-orbit interaction [10]. The sign of the coefficient $D$ generally depends on the detailed band structure [10], while Ye *et al.* argued that $\tilde{D}$ should be positive [4]. In any case, a crucial observation here is that the weak chiral symmetry-breaking term (5) guarantees a net total chirality to be induced if the sample is magnetized. Net magnetization may be generated spontaneously (ferromagnet or reentrant SG) or induced by applying external fields.

I now go on to analyze the behavior of the anomalous Hall resistivity of SG based on Eqs. (4) and (5). I assume for the time being that the system does not possess a spontaneous magnetization; namely, the magnetization is the one induced by external magnetic field $H$.

The quantities playing a crucial role in the following analysis are the linear and nonlinear *chiral* susceptibilities, defined by

$$X_\chi = \frac{d\chi_0}{dH_\chi}\bigg|_{H_\chi=0}, \qquad X_\chi^{nl} = \frac{1}{6}\frac{d^3\chi_0}{dH_\chi^3}\bigg|_{H_\chi=0}, \tag{6}$$

where $H_\chi$ is the "chiral field" conjugate to the net chirality $\chi_0$, i.e., $H_\chi$ couples to the net chirality as $-H_\chi\chi_0$ in the spin Hamiltonian. Note that the chiral symmetry-breaking interaction discussed above, Eq. (5), has exactly this form with $H_\chi = -\tilde{D}M$.

With use of the linear and nonlinear chiral susceptibilities, the total chirality can be written as

$$\chi_0 = -X_\chi(\tilde{D}M) - X_\chi^{nl}(\tilde{D}M)^3 + \cdots. \tag{7}$$

If one substitutes this into Eq. (4), one gets the chirality contribution to the anomalous part of the Hall resistivity as

$$\rho_{xy}^{(\mathrm{chiral})} = -\tilde{C}\tilde{D}M[X_\chi + X_\chi^{nl}(\tilde{D}M)^2 + \cdots]. \tag{8}$$

Including the contributions of the skew scattering and the side jump, the Hall coefficient $R_s$, defined as the anomalous Hall resistivity divided by the magnetization $R_s = \rho_{xy}/M$, is given by

$$R_s = -\tilde{A}\rho - \tilde{B}\rho^2 - \tilde{C}\tilde{D}[X_\chi + X_\chi^{nl}(\tilde{D}M)^2 + \cdots]. \quad (9)$$

The total Hall resistivity contains in addition the contribution from the normal part, which, we assume throughout this analysis, has properly been subtracted. One can immediately see from Eq. (9) that the anomalous Hall coefficient $R_s$ carries information of the chiral susceptibilities. In particular, in the linear regime where the magnetization is sufficiently small and the Hall resistivity is proportional to $M$, the chirality contribution to $R_s$ is proportional to the linear chiral susceptibility $X_\chi$.

In the standard measurements of Hall resistivity, the dc magnetic field is applied, either in field-cooling (FC) or zero-field-cooling (ZFC) conditions. As is well known, at the SG transition temperature $T = T_g$, the linear magnetic susceptibility $X_m = [(dM)/(dH)]_{H=0}$ (to be distinguished from the linear chiral susceptibility) exhibits a cusp accompanied by the onset of deviation between the FC and ZFC susceptibilities [1]. Sharp cusp of the linear susceptibility at $T = T_g$ is known to be rounded off by applying weak external magnetic fields, which is also manifested in the well-known negative divergence of the nonlinear magnetic susceptibility $X_m^{nl} = (1/6)[(d^3M)/(dH^3)]_{H=0}$ at $T = T_g$[1]. By contrast, the resistivity $\rho$ of canonical SG exhibits no detectable anomaly at $T = T_g$[1].

The Hall resistivity is generally given by the combination of both the magnetic and chiral susceptibilities, as seen from Eq. (8) with $M = X_mH + X_m^{nl}H^3 + \cdots$. By contrast, one can extract information solely about the chiral susceptibilities from the Hall coefficient $R_s$ which is obtained by dividing the Hall resistivity by the magnetization measured simultaneously or in the same condition. Here, note that the magnetization of SG exhibits a singular behavior at $T = T_g$. Furthermore, by examining the $M$ dependence of the Hall coefficient in the nonlinear regime, one can extract information about the nonlinear chiral susceptibility. Anyway, in contrast to the standard magnetic susceptibilities measurable by the conventional technique, information about the chiral susceptibilities have thus far been hard to get experimentally and, if measurable as above, would be very valuable.

Next, I wish to give predictions on the behavior of the anomalous Hall coefficient based on the chirality scenario of SG transition [2,3]. The chirality scenario predicts that, in both isotropic and weakly anisotropic Heisenberg SGs, the chirality behaves as an order parameter of the transition (chiral-glass transition). The singular part of the free energy should satisfy the scaling form,

$$f_s \approx |t|^{2\beta_\chi+\gamma_\chi} F_\pm\left(\frac{H_\chi^2}{|t|^{\beta_\chi+\gamma_\chi}}\right), \quad (10)$$

where $\beta_\chi$ and $\gamma_\chi$ are the chiral-glass order parameter and chiral-glass susceptibility exponents, respectively, $t \equiv (T - T_g)/T_g$ is a reduced temperature, and $F_\pm(x)$ is a scaling function either above ( + ) and below ( − ) $T_g$. Numerical estimates give $\beta_\chi \sim 1$ and $\gamma_\chi \sim 2$ [14,15]. By differentiating Eq. (10) with respect to $H_\chi$ and putting $H_\chi = 0$, one sees that at $T = T_g$ the linear chiral susceptibility exhibits a cusplike singularity while the nonlinear chiral susceptibility exhibits a negative divergence,

$$X_\chi \approx c_0^{(\pm)}|t|^{\beta_\chi} + b_0(t), \qquad X_\chi^{nl} \approx c_2^{(\pm)}|t|^{-\gamma_\chi} + b_2(t), \quad (11)$$

with $\beta_\chi \sim 1$ and $\gamma_\chi \sim 2$, where $c_0^{(\pm)} < 0$ and $c_2^{(\pm)} < 0$ are constants describing either above or below $T_g$, while $b_0(t) > 0$ and $b_2(t)$ represent regular terms coming from the nonsingular part. Concerning the standard magnetic susceptibilities, the chirality scenario predicts that, in the realistic case of weakly anisotropic Heisenberg SG, $X_m$ and $X_m^{nl}$ exhibit the same singularities as $X_\chi$ and $X_\chi^{nl}$, which are caused by the spin-chirality recoupling due to the random magnetic anisotropy. (In the hypothetical limit of zero anisotropy, because of the spin-chirality decoupling in the isotropic system, $X_m$ and $X_m^{nl}$ are predicted to exhibit less singular behaviors very different from those of $X_\chi$ and $X_\chi^{nl}$. But, after all, a certain amount of anisotropy is inevitable in real SG, which eventually causes the spin-chirality recoupling.)

The anomalous Hall coefficient of SG should be dominated by the singular behaviors of the chiral susceptibilities, since the first and second terms of the right-hand side of Eq. (9) can be regarded as a regular background because of the nonsingular behavior of $\rho$. By combining the observations above, the following predictions follow. (i) The linear part of $R_s$, which is $R_s$ itself in the linear regime where $R_s$ is proportional to the magnetization $M$, exhibits a cusplike anomaly at $T = T_g$, possibly accompanied by the onset of the deviation between the FC and ZFC results. This cusplike singularity is rounded off in the presence of a finite magnetization. (ii) The nonlinear part of $R_s$, which can be extracted by examining the $M$ dependence of $R_s$ in the nonlinear regime, exhibits a divergence at $T = T_g$ characterized by the exponent $\gamma_\chi \sim 2$, which is equal to the standard nonlinear susceptibility exponent $\gamma$. (iii) The chiral part of $R_s$, obtained by properly subtracting the background due to the possible contribution of the skew scattering and the side jump, etc., is expected to obey the scaling form,

$$R_s^{(\mathrm{chiral})} \approx |t|^{\beta_\chi} G_\pm\left(\frac{M^2}{|t|^{\beta_\chi+\gamma_\chi}}\right), \quad (12)$$

where $G_\pm(x)$ is a scaling function either above or below $T_g$. The subtraction of the background might be performed by analyzing the temperature dependence of $R_s$ based on Eq. (9), using the data of the resistivity $\rho$. (iv) The sign of the Hall resistivity depends on the signs and the relative magnitudes of constants $\tilde{A}$, $\tilde{B}$, $\tilde{C}$, and $\tilde{D}$ ($X_\chi$ is positive by definition). Hence, the sign of $\rho_{xy}$ seems nonuniversal, depending on the band structure of the material. In cases where the single-band approximation and the naive argument of Ref. [4] concerning the sign of $\tilde{D}$ are valid, one has $\tilde{C} > 0$ (with $J_{sd} > 0$) and $\tilde{D} > 0$, which means $\rho_{xy}^{(\mathrm{chiral})}$ is negative in canonical SG. This seems consistent with experiment [16,17]. The cusplike singularity was observed there at least in the Hall resistivity [16,17], consistent with the present result.

Note that even the conventional mechanism of the anomalous Hall effect (the skew-scattering or the side-jump mechanism) predicts that the Hall resistivity exhibits a cusplike singularity at $T = T_g$, which is a reflection of the cusplike singularity of the magnetic susceptibility. However, the conventional mechanism also predicts that the Hall coefficient $R_s$ behaves in a nonsingular manner at $T = T_g$ as $\rho$ and $\rho^2$. A highly nontrivial issue is then whether the Hall coefficient, not just the Hall resistivity, exhibits an anomaly at $T = T_g$. If singular behavior is observed in $R_s$, it is likely to be of chirality origin.

One can give a rough order estimate of $\rho_{xy}^{(\mathrm{chiral})}$. In typical canonical SG such as AuFe and CuMn, $J_{sd}/\epsilon_F$ and $J\tau$ are of order $10^{-1}$ and $10^0$, respectively. Then, from Eq. (2), $\rho_{xy}^{(\mathrm{chiral})}$ is estimated to be of order $M\chi_0$ in units of $\rho_0$. Since the magnitude of the chiral symmetry-breaking interaction (5) is of the order of the DM interaction, the induced chirality $\chi_0$ is of the order of [DM interaction]/[RKKY interaction]. This ratio is a material dependent parameter, being small for CuMn, for example, $10^{-2}$, and relatively large for AuFe, for example, $10^{-1}$ or more. Thus, if the sample is magnetized 10% of the saturation value, one expects for AuFe $\rho_{xy}^{(\mathrm{chiral})}$ of order percents of $\rho_0$ or even more. For more isotropic materials such as CuMn and AgMn, the chiral contribution would be reduced being proportional to the strength of the DM interaction.

Finally, I wish to discuss the reentrant SG with a spontaneous magnetization. With decreasing temperature, the reentrant SG exhibits successive transitions, first from para to ferro at $T = T_c$, then from ferro to reentrant SG at $T = T_g$. The present result for the Hall resistivity also applies to such reentrant SG around $T = T_g$, only if $M$ is treated as including the spontaneous magnetization. Below $T = T_g$, an additional contribution from the chiral order sets in, giving rise to a cusp in the Hall resistivity at $T = T_g$. As often observed under FC conditions, magnetization of reentrant SG is saturated at temperatures far above $T_g$, with very little anomaly at $T = T_g$. In such a case, if anomaly is observed in the FC mode in the Hall resistivity at $T = T_g$, this can be identified as arising from the chirality. Indeed, a cusplike anomaly at $T = T_g$ was recently observed in manganite reentrant SG $La_{1.2}Sr_{1.8}Mn_2O_7$ [18] and in reentrant SG alloy $Fe_{1-x}Al_x$ [19], suggesting that the observed anomaly is of chirality origin.

In summary, I examined the Hall resistivity of canonical SG, and have found that the Hall coefficient gives information about the linear and nonlinear chiral susceptibilities of SG. Based on the chirality scenario, predictions were given on the behavior of the Hall coefficient of canonical SG. I hope the present work will stimulate further experimental activities on the chiral order and the Hall resistivity of SG and related materials.

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[1] For reviews on spin glasses, see, e.g., K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986); K. H. Fischer and J. A. Hertz, *Spin Glasses* (Cambridge University Press, Cambridge, England, 1991); J. A. Mydosh, *Spin Glasses* (Taylor & Francis, London, 1993); *Spin Glasses and Random Fields*, edited by A. P. Young (World Scientific, Singapore, 1997).
[2] H. Kawamura, Phys. Rev. Lett. **68**, 3785 (1992).
[3] H. Kawamura, Phys. Rev. Lett. **80**, 5421 (1998).
[4] J. Ye, Y. B. Kim, A. J. Millis, B. I. Shraiman, P. Majumdar, and Z. Tesanovic, Phys. Rev. Lett. **83**, 3737 (1999).
[5] S. H. Chun, M. B. Salamon, Y. Lyanda-Geller, P. M. Goldbart, and P. D. Han, Phys. Rev. Lett. **84**, 757 (2000).
[6] K. Ohgushi, S. Murakami, and N. Nagaosa, Phys. Rev. B **62**, R6065 (2000).
[7] Y. Taguchi and Y. Tokura, Phys. Rev. B **60**, 10280 (1999); Europhys. Lett. **54**, 401 (2001).
[8] Y. Taguchi, Y. Oohara, H. Yoshizawa, N. Nagaosa, and Y. Tokura, Science **291**, 2573 (2001).
[9] S. Yoshii *et al.*, J. Phys. Soc. Jpn. **69**, 3777 (2000); S. Iikubo *et al.*, J. Phys. Soc. Jpn. **70**, 212 (2001); Y. Yasui *et al.*, J. Phys. Soc. Jpn. **70**, 284 (2001); T. Kageyama *et al.*, J. Phys. Soc. Jpn. **70**, 3006 (2001); Y. Yasui *et al.*, cond-mat/0211369.
[10] G. Tatara and H. Kawamura, J. Phys. Soc. Jpn. **71**, 2613 (2002).
[11] R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
[12] J. Smit, Physica (Utrecht) **21**, 877 (1955); **24**, 39 (1958).
[13] J. M. Luttinger, Phys. Rev. **112**, 739 (1958).
[14] K. Hukushima and H. Kawamura, Phys. Rev. E **61**, R1008 (2000).
[15] H. Kawamura and D. Imagawa, Phys. Rev. Lett. **87**, 207203 (2001); D. Imagawa and H. Kawamura, J. Phys. Soc. Jpn. **71**, 127 (2002).
[16] S. P. McAlister and C. M. Hurd, Phys. Rev. Lett. **37**, 1017 (1976); J. Phys. F **8**, 239 (1978).
[17] R. D. Barnard and I. Ul-haq, J. Phys. F **18**, 1253 (1988).
[18] S. H. Chung, Y. Lyanda-Geller, M. B. Salamon, S. Suryanarayanan, G. Dhalenne, and A. Revcolevschi, J. Appl. Phys. **90**, 6307 (2001). Here $J_{sd}$ is probably negative (double exchange), suggesting $\tilde{C} < 0$. The experimental data given in Fig. 3b of this reference might be understandable with $\tilde{C} < 0$ and $\tilde{D} > 0$, if the conventional contribution gives a negative background, i.e., $\tilde{A} > 0$ and/or $\tilde{B} > 0$.
[19] T. Kageyama *et al.*, cond-mat/0211368.