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Chiral susceptibility of canonical spin glasses from Hall effect measurements

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Abstract

The extraordinary Hall resistivity ρ_{ex} and the magnetization M of canonical spin glasses were measured simultaneously as a function of temperature, with close attention to thermal and magnetic field hysteresis. The data for ρ_{ex} show an anomaly at the spin glass transition temperature T_g. Moreover, the value of ρ_{ex}/M, which represents the chiral susceptibility of the system in the present case, also shows the anomaly. In conventional theories, the extraordinary Hall resistivity ρ_{ex} is represented, $\rho_{ex} = M(A\rho + B\rho^2)$, where ρ is the resistivity, and A and B are constants. Since $\rho(T)$ is monotonic and smooth, the behaviour of ρ_{ex}/M clearly indicates that one has to include another term in the expression for ρ_{ex}. The critical phenomena of the spin glass transition from Hall effect measurements is discussed in comparison with that from magnetization measurements. One of the critical exponents for the chirality, δ_χ, was obtained from the field dependence of R_S. The value of δ_χ should be compared with that of δ determined from the magnetization measurements. The results can be interpreted consistently in terms of a chirality ordering model of canonical spin glasses.

For the last several decades, spin glass (SG) has been extensively studied as a prototype of complex systems [1]. There is a consensus that the SG transition is a 'true' thermodynamic phase transition. The most familiar and well-studied SG systems are the dilute magnetic alloys such as AuFe, AgMn and CuMn, so-called canonical SGs. In canonical SGs, the localized moments of randomly distributed magnetic atoms interact with each other via the s–d exchange interaction mediated by the conduction electrons, the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. The RKKY interaction is isotropic, and in the absence of spin anisotropy a canonical SG is expected to be well described by the three-dimensional (3D) Heisenberg model. In real alloys the Dzyaloshinsky–Moriya (DM) random anisotropy is inevitably present, whose magnitude depends on the non-magnetic host metal. In many cases the experimental results of canonical SGs have been interpreted by the mean field model which is an extended Sherrington–Kirkpatrick (SK) model of a Heisenberg spin system [1]. The theoretical arguments still continue whether or not a 3D Heisenberg random spin system can show an SG transition without additional anisotropy at a finite temperature [2, 3]. However, these theoretical works including the mean field theory face serious difficulties in comparing with the experimental results even when the DM anisotropy term is taken into consideration.

In many theories, T_g depends on only the magnitude of the anisotropy D in small D region [2]. Though the value of D of AuFe is about ten times larger than that of CuMn, these alloys with the same concentration of magnetic impurities have almost the same SG transition temperatures. The discrepancy between the experimental results and the mean field theory has been pointed out on the SG transition line in a magnetic field. The Almeida–Thouless (AT)-like line, $H \sim A(T_g - T_f(H))^{3/2}$, is observed in most canonical SGs, but the coefficient A is a few tens of times smaller than that predicted by the mean field theory [4]. There are contradictions in the critical phenomena of the SG transition. The Heisenberg–Ising crossover [5], which is expected for a 3D Heisenberg SG with DM anisotropy, has not been clearly observed yet. The scaling analysis in the appropriate temperature and magnetic field regions has given the same critical exponents to AuFe and AgMn even though they have quite different magnitudes of the anisotropy [6–8]. This suggests that the family of canonical SGs belongs to the same universality class.

Kawamura [9] proposed the chirality hypothesis, which can overcome the above-mentioned difficulties. The scenario of SG transition by the chirality mechanism is as follows: an isotropic Heisenberg random spin system does not undergo an SG transition by itself but has a 'chiral glass' (CG) transition at a finite temperature. The (scalar) chirality, $\chi_{ijk} \equiv \vec{S}_i \cdot \vec{S}_j \times \vec{S}_k$, is not coupled to the spin so long as the isotropy is perfect. Then, however, the possible weak random DM anisotropy can mix the spin with the chirality. Consequently, an apparent SG transition becomes observable at a finite temperature in a real spin system. Numerical estimates [10] give the critical exponents $\beta_\chi \sim 1, \gamma_\chi \sim 2$ to the chiral glass transition. These values are similar to the corresponding critical exponents of the SG transition of canonical SGs deduced from nonlinear susceptibility measurements [6–8]. Though the chirality scenario is attractive, experimental testing has not progressed because of the difficulty in direct measurement of the chirality.

Recently, Tatara and Kawamura [11] have derived the chirality contribution to the extraordinary Hall resistivity by applying the linear response theory and the perturbation expansion to the weak coupling s–d Hamiltonian. Kawamura [12] has made predictions on the behaviour of the extraordinary Hall resistivity of canonical SGs based on the chirality scenario of the SG transition. The main purpose of the present article is to verify the chirality scenario by simultaneously measuring the Hall resistivity and the magnetization in canonical SGs.

The samples used for the measurements are AuFe and AuMn alloys. We prepared ingots of the sample alloys by melting constituent elements in an argon arc-furnace. A 'cloverleaf-shaped' sample 6 mm in diameter and 0.2 mm thick was cut out by a spark cutting machine from each of the ingots. The sample was sealed in a vacuous quartz ampoule, annealed at 850 °C for 1 week, and quenched to the room temperature. It should be noted that all measurements of the Hall resistivity, the magnetization and the resistivity were done on the same sample.

Several authors have reported on the Hall effect of canonical SGs [13–15]. The temperature dependence of Hall resistivity ρ_H is basically similar to that of the AC susceptibility or the zero-field cooled (ZFC) magnetization under a weak magnetic field. In the conventional method of the Hall measurement, a transverse voltage is measured with a constant current in an applied magnetic field perpendicular to both the direction of the current flow and the voltage drop. Practically, the terminal misalignment or the gradient of potential surface produces a spurious Hall voltage. In order to cancel this spurious Hall voltage, the magnetic field or the sample is usually flipped through 180°, and the mean value of voltage drop is adopted as the 'true' Hall voltage. All the previous Hall effect measurements of canonical SGs [13–15] were made by using this procedure. As is pointed out by the author of [13], the flipping of the sample in a field breaks the thermodynamic state of the SG. It is well known that the thermodynamic state of an SG is strongly dependent on the field and the temperature hysteresis. Thus, the above-mentioned procedure cannot be adopted in the present experiments. Accordingly, we have developed a simultaneous measurement system of ρ_H and M under the correct ZFC and field-cooled (FC) conditions. All the measurements of ρ_H, M and the resistivity ρ were made while the sample was embedded in a commercial-type SQUID magnetometer MPMS-7 (Quantum Design). The Hall resistivity and the resistivity measurements were done on the cloverleaf-shaped sample with four terminals following the van der Pauw method [16]. The advantage of this method is that one can obtain the Hall resistivity and the resistivity on the same sample by changing the combination of the terminals, that is, the flipping of the sample is not necessary. The actual measurement procedure is described below. In the absence of the field, the transverse voltage $V_{xy}(0)$ to the current flow is recorded and the Hall voltage $V_H(H)$ is obtained by subtraction: $V_{xy}(H) - V_{xy}(0)$. The residual field of the MPMS magnet was estimated to be less than 1.6 G. The effect of the residual field of this magnitude is negligibly small in the present work. In the ZFC measurements, the sample is cooled in zero field to 5 from 60 K. After a field is applied, $V_{xy}(H)$ and M are simultaneously measured at constant temperature increments of 1 K. The FC measurements are successively made in the same way as the ZFC measurements after cooling the sample in the field. Consequently ρ_H and M can be obtained under the ZFC and FC conditions in the same thermal and field conditions without flipping the sample in the field. The temperature and field are controlled by using the MPMS sequence system. Though ρ under the ZFC and FC conditions is separately measured from ρ_H and M, the same sequence ensure the same experimental conditions. Since the Hall signal is very small in the present case, we use a lock-in amplifier with a highly stable AC constant current source.

Recently, some features [17–20] of the extraordinary Hall coefficient $R_s \equiv \rho_{ex}/M$, which are not understood by the conventional theory [21, 22], have been reported in SGs; ρ_{ex} is the extraordinary Hall resistivity and M is the magnetization, Also, in [18–20], a deviation between the ZFC and the FC data below T_g has been reported.

Figure 1 shows one of the results of the simultaneous measurement of ρ_H and M. For the ZFC result, the sign and the magnitude of ρ_H are consistent with those of the previous measurement [13]. ρ_H in the FC condition was obtained for the first time in this measurement. In the figure, one can see that the temperature dependence of ρ_H is quite similar to that of M, and that significant differences between ZFC and FC results of ρ_H appear below the temperature $T_g(H)$ where the difference also appears in M.

The Hall resistivity of magnetic materials consists of two parts: the ordinary part ρ_o and the extraordinary part ρ_{ex}. Extrapolations to high temperatures to obtain an estimate of the ordinary part for the present alloy indicate that the ordinary Hall coefficient is about $-8 \times 10^{-13}\ \Omega$ cm G^{-1}. In the temperature and the field ranges of interest of the present system, the ordinary part is much smaller than the extraordinary part; therefore, we hereafter neglect the ordinary part, $\rho_H = \rho_o + \rho_{ex} \sim \rho_{ex}$.

Figure 2 shows the temperature dependence of the Hall resistivity ρ_{ex} divided by the simultaneously measured M in the fields indicated. One can see that the ZFC curve at 500 G shows a maximum around $T_g(H)$ and that the maximum is suppressed by the field. It is remarkable that the value of ρ_{ex}/M also has the differences between ZFC and FC measurements below $T_g(H)$. $T_g(H)$ shifts to lower temperatures as the field increases. It is well known that the SG order is sensitive to a magnetic field and the magnetic susceptibility involves large

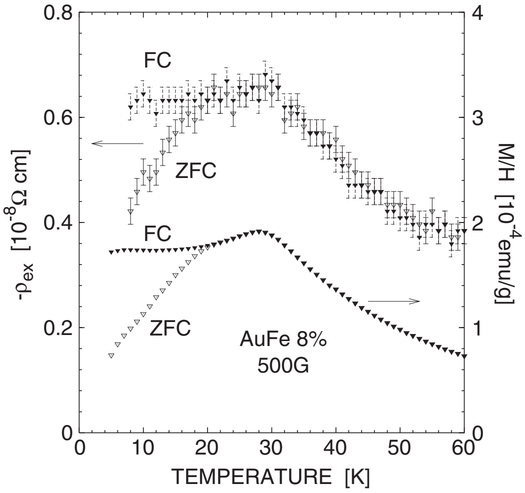

Figure 1. Simultaneous measurement of the Hall resistivity ρ_H ($\sim\rho_{ex}$) and magnetization M for AuFe 8 at.% Fe (taken from [18]).

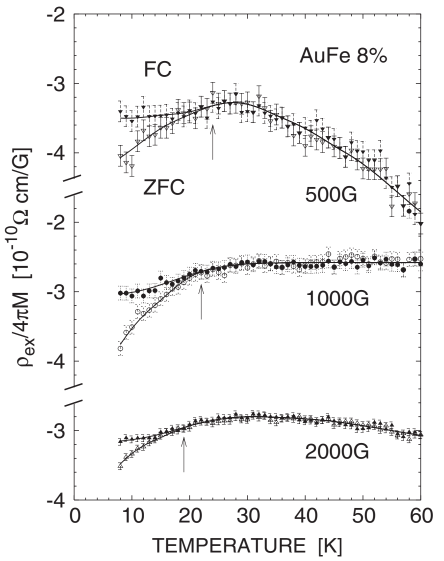

Figure 2. Temperature dependence of ρ_{ex}/M in the fields indicated. The arrows mark $T_g(H)$ (taken from [18]).

nonlinear terms. The large magnetic field dependence of ρ_{ex}/M around T_g has been predicted in the chirality scenario of the canonical SGs [12]. In order to discuss this point in more detail, precise measurements under smaller fields are required.

In the conventional theories [21, 22] the extraordinary Hall resistivity is represented as

$$\rho_{ex} = M(A\rho + B\rho^2), \tag{1}$$

where ρ is the resistivity, and A and B are constants relevant to the detailed band structure of the conduction electrons. In equation (1), the first and the second terms represent the skew scattering and the side jump effect respectively. In the present case, the temperature dependence

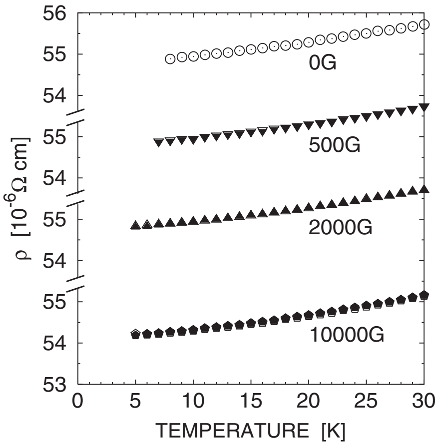

Figure 3. Temperature dependence of resistivity ρ under ZFC and FC conditions in the fields indicated (taken from [18]).

of ρ, as shown in figure 3, is monotonic and smooth even around $T_g(H)$, and the difference between ZFC and FC is not observed in any field. The measurements of ρ were done with the same sequence used for the simultaneous measurements of the ρ_{ex} and the M. Together with this monotonic $\rho(T)$, the behaviour of ρ_{ex}/M in figure 2 clearly indicates that one has to include another term in equation (1).

Tatara and Kawamura [11] have shown, using the standard s–d Hamiltonian, that an additional term of the extraordinary Hall effect appears when the total chirality $\chi_0 \neq 0$. The total chirality χ_0 is the sum of the local chirality χ_{ijk} weighted by the geometrical factor which depends on the distance between the spins. The contribution of the total chirality to the extraordinary Hall effect is independent of those of the conventional ones. Then, the extraordinary Hall resistivity is expressed as follows [11]:

$$\rho_{ex} = M(A\rho + B\rho^2) + C\chi_0. \tag{2}$$

Since Heisenberg spins are frozen in a spatially random manner in the SG ordered state, the sign of the local chirality χ_{ijk} appears randomly, which inevitably leads to the vanishing of the total chirality, $\chi_0 = 0$. Therefore, the chirality-driven extraordinary Hall effect vanishes in bulk SG samples. One possible mechanism to realize a finite uniform chirality was proposed for the strong coupling case by Ye *et al* [23]. The authors showed that the spin–orbit interaction in the presence of a net magnetization M contains a term $H_{so} = DM\chi_0$. In terms of this Hamiltonian, they have explained the extraordinary Hall effect of colossal magnetoresistance manganites. This idea was applied to the weak coupling system by Tatara and Kawamura in a perturbation calculation [11]. They have shown that ρ_{ex}/M is expressed as follows:

$$\frac{\rho_{ex}}{M} = A\rho + B\rho^2 + X_\chi + X_\chi^{nl} M^2 + \cdots, \tag{3}$$

where X_χ and X_χ^{nl} are constants with respect to M. The above argument contains two physically important meanings. First, ρ_{ex}/M no longer depends on M when M is sufficiently small. Because $\rho(T)$ is monotonic, the temperature dependence of observed ρ_{ex}/M should be explained in terms of the temperature dependence of the chiral contribution terms. Second, the fact that the uniform chirality χ_0 is induced, through H_{so}, by the uniform magnetization M means that M acts as a 'symmetry-breaking field' of χ_0 [11, 12]. Therefore, M is regarded as a 'chiral field', and the constants X_χ and X_χ^{nl} in equation (3) are a 'chiral linear susceptibility'

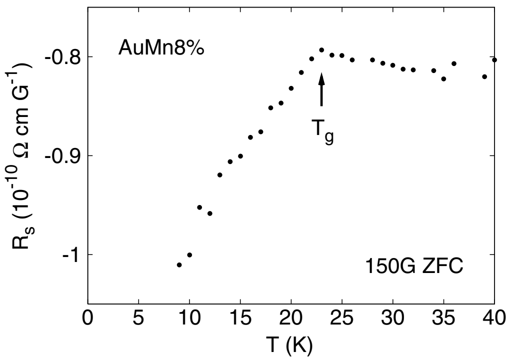

Figure 4. Temperature dependence of ZFC R_S around $T_g = 24.3$ K. $R_S(T)$ was measured under 150 G.

and a 'chiral nonlinear susceptibility', respectively. In the case of a ferromagnetic transition, the order parameter is a spontaneous magnetization M, and the symmetry-breaking field is a uniform magnetic field H. The order parameter susceptibility, which is the order parameter divided by the conjugate field, namely the uniform magnetic field, shows a strong anomaly at the transition point. According to the chiral scenario of the canonical SG, the uniform chiral susceptibility must show a 'cusp' at the transition temperature and also ZFC and FC hysteresis [12], as exactly evidenced experimentally in figure 2.

In the experiments on AuFe, a difference between ZFC and FC measurements below the spin glass transition temperature T_g was observed in the temperature dependence of the anomalous Hall coefficient $R_S(T)$ obtained from the simultaneous measurements of the magnetization M and the anomalous Hall resistivity ρ_{ex}, while the resistivity $\rho(T)$ was monotonic and smooth, and a difference between ZFC and FC measurements was not observed. Since R_S is just a function of ρ in the conventional theory [21, 22], another contribution has to be included in R_S. The origin of the contribution is probably the chirality which randomly orders in the system. The amplitude of the minimum field was 2000 G due to the sensitivity of the measurement system [18]. This amplitude of the field is not appropriate to investigate the critical phenomena of the spin glass transition. We improved the sensitivity of the measurements and we were able to study the critical phenomena from Hall effect measurements.

The spin glass transition temperature $T_g = 24.3$ K of a AuMn spin glass sample with 8 at.% Mn was determined by the cusp temperature in the low-field magnetization measurement. Figure 4 shows the temperature dependence of ZFC R_S of the sample under a field of 150 G which is one decade smaller than that of the experiments on AuFe mentioned above. The sharp 'cusp' in ZFC $R_S(T)$ at T_g is easily suppressed by magnetic fields. The critical exponent δ_χ related to the conjugate field of the order parameter is obtained from this suppression of $R_S(T_g)$. Since the sign of the local chirality appears randomly in this case, the conjugate field is proportional to M^2 [12]. Thus the critical exponent δ_χ can be obtained from the magnetization dependence of $R_S(M(H))$ at T_g.

Figure 5 shows the plot of $\Delta R_S(M^2)$ at T_g in a log scale, which is derived from the field dependence of R_S, where $\Delta R_S \equiv R_{S0} - R_S$ and R_{S0} is the extrapolation value to $M = 0$. The extraordinary Hall coefficient including the conventional term is derived as follows:

$$R_S \equiv \rho_{ex}/M = A\rho + B\rho^2 + CX_\chi + C'X_\chi^{nl}M^2 + \cdots,$$

where A, B, C and C' are constants and X_χ and X_χ^{nl} are linear and nonlinear chiral

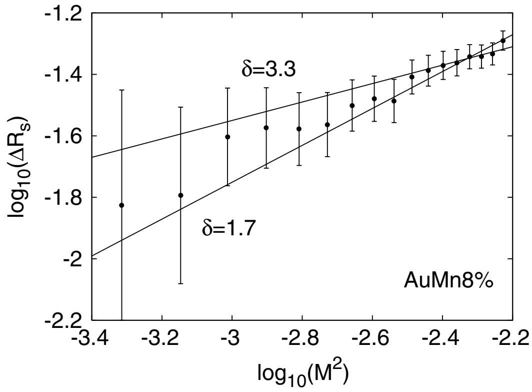

Figure 5. Log–log plot of ΔR_S versus M^2 at T_g. The two straight lines express $\delta_\chi = 1.7$ and $\delta_\chi = 3.3$ respectively.

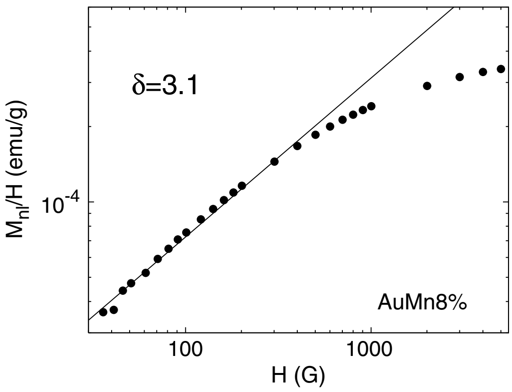

Figure 6. Log–log plot of M_{nl} versus H at T_g. The straight line expresses $\delta = 3.1$.

susceptibilities respectively. Since $R_{S0} = (\rho_{ex}/M)_{M\to 0}$ becomes $A\rho + B\rho^2 + CX_\chi$, ΔR_S is proportional to $X_\chi^{nl} M^2$ in the regime where M is sufficiently small. If the 'true' order parameter of the spin glass transition is not the spin but the chirality, ΔR_S should satisfy the power law at T_g; $\Delta R_S \sim (M^2)^{1/\delta_\chi}$. The two straight lines in figure 5 show the behaviour with $\delta_\chi = 1.7$ and $\delta_\chi = 3.3$ respectively, and δ_χ is determined to be 2.5 ± 0.8. This value of δ_χ should be compared with that of δ determined from the magnetization measurements, where δ is defined as $q \sim (H^2)^{1/\delta}$ at T_g and q is the Edwards–Anderson order parameter.

The critical exponents of the spin glass transition of canonical spin glass systems were extensively investigated [6–8] from the nonlinear magnetization measurements. The value of $\delta = 3.0$ determined in AgMn and AuFe systems is almost the same as that in the AuMn system shown in figure 6. The details of the scaling analysis of the nonlinear magnetization will be discussed elsewhere [20]. It should be noted that the value of δ_χ coincides with that of δ within experimental errors. The critical phenomena of the spin glass transition of the AuMn system from the Hall effect measurements are compatible with the predictions of the chirality scenario of the spin glass transition [12].

In summary, simultaneous measurements of the extraordinary Hall resistivity and the magnetization were carried out on canonical spin glass systems AuFe and AuMn. The temperature dependence of ρ_{ex}/M, which represents the uniform chiral susceptibility, shows a cusp at T_g and ZFC and FC hysteresis below T_g. The critical exponent δ_χ of AuMn was obtained from the M-dependence of the Hall resistivity at T_g. The value coincides with that from the magnetization measurement within experimental errors. These observations are compatible with the prediction by the Kawamura chirality scenario of canonical SGs [12].

Acknowledgments

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