Direct Observation of Chiral Susceptibility in the Canonical Spin Glass AuFe

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The extraordinary Hall resistivity ρ_{xy} and the magnetization M of a canonical spin glass AuFe (8 at.% Fe) were measured simultaneously as functions of temperature with the best care to the thermal and the magnetic field hysteresis. The data of ρ_{xy} show an anomaly at the spin glass transition temperature T_g and have different zero field cooling (ZFC) and field cooling (FC) measurements below T_g. Moreover, the value of ρ_{xy}/M, which represents the chiral susceptibility of the system in the present case, also shows the difference between ZFC and FC measurements. The results are consistent with the predictions of the chirality scenario of canonical spin glasses by Kawamura.

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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 SG. 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 the 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 nonmagnetic host metal. In many cases the experimental results of canonical SGs have been interpreted by the mean field model which is an extended SK model of Heisenberg spin system [1]. The theoretical arguments still continue whether or not a 3D Heisenberg random spin system can show 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 the small D region [2]. Though the D of AuFe is about 10 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 about 20 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 the 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 [6,7]. This suggests that the family of the canonical SGs belongs to the same universality class.

Kawamura [8] proposed the chirality hypothesis, which can overcome above mentioned difficulties. The scenario of SG transition by the chirality mechanism is as follows: an isotropic Heisenberg random spin system does not undergo SG transition by itself but has "chiral glass" 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 as far 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 [9] give the critical exponents of $\beta_\chi \sim 1$, $\gamma_\chi \sim 2$, to the chiral glass transition. These values are similar to the corresponding critical exponents of the SG transition of canonical SGs deduced from the nonlinear susceptibility measurements [6,7]. Though the chirality scenario is attractive, the experimental test has not progressed because of the difficulty in the direct measurement of the chirality.

Recently Tatara and Kawamura [10] 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 [11] has made predictions on the behavior of the Hall resistivity of canonical SGs based on the chirality scenario of SG transition. The main purpose of the present article is to verify the chirality scenario by simultaneously measuring the Hall resistivity ρ_{xy}, the magnetization M, and the resistivity ρ in canonical SG.

The sample used for the measurements is AuFe 8 at. % Fe. We prepared an ingot of the sample alloy 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. The sample was sealed in a vacuous quartz ampoule and annealed at 850 °C for one week and quenched to room temperature. It should be noted that all measurements of ρ_{xy}, M, and ρ were done on the same sample.

Several authors have reported on the Hall effect of canonical SGs [12–14]. The temperature dependence of Hall resistivity is basically similar to that of the ac susceptibility or the zero field cooling (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 [12–14] were made by taking this procedure. As is pointed out by the author of Ref. [12], the flipping of the sample in a field breaks the thermodynamic state of the SG. It is well known that the thermodynamic state of 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_{xy} and M under the correct ZFC and FC condition. All the measurements of ρ_{xy}, M, and 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 [15]. 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_{xy} and M can be obtained under the ZFC and FC conditions in the same thermal and field condition 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 the ρ_{xy} and M, the same sequence ensure the same experimental condition. 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.

Figure 1 shows one of the results of the simultaneous measurement of ρ_{xy} and M. For the ZFC result, the sign and the magnitude of ρ_{xy} are consistent with those of the previous measurement [12]. The ρ_{xy} 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_{xy} is quite similar to that of M, and that significant differences between ZFC and FC results of ρ_{xy} appear below the temperature $T_f(H)$ where the difference also appears in M.

The Hall resistivity of magnetic materials consists of two parts, the ordinary part and the extraordinary part. Extrapolations to high temperatures to obtain an estimate of the ordinary part for the present alloy indicate that the ordinary Hall coefficient is about -8×10^{-13} Ω 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.

Figure 2 shows the temperature dependence of the Hall resistivity ρ_{xy} divided by the simultaneously measured M in the fields indicated. One can see that the ZFC curve at 500 G shows a maximum around $T_f(H)$ and that the maximum is suppressed by the field. It is remarkable that the value of ρ_{xy}/M also has the differences between ZFC and FC below $T_f(H)$ [16]. The $T_f(H)$ 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 nonlinear terms. The large magnetic field dependence of ρ_{xy}/M around T_g has been predicted in the chirality scenario of the canonical SG [11]. In order to discuss this point in more detail, precise measurements under smaller fields are required.

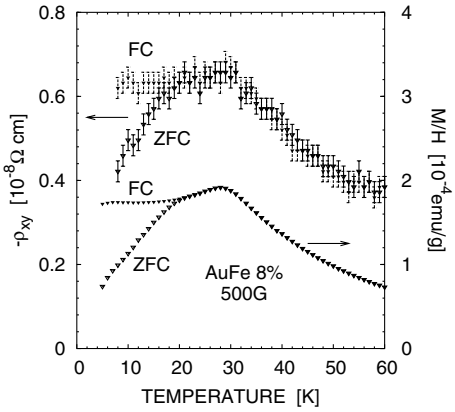

FIG. 1. Simultaneous measurement of the Hall resistivity ρ_{xy} and magnetization M for AuFe 8 at. % Fe.

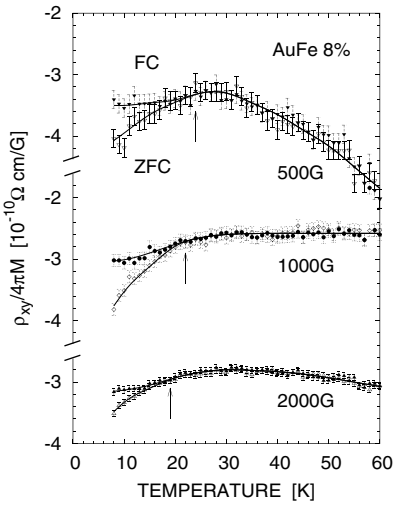

FIG. 2. Temperature dependence of ρ_{xy}/M in the fields indicated. The arrows mark $T_f(H)$.

In the conventional theories [17,18] the extraordinary Hall resistivity is represented,

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

where ρ is the resistivity, A and B are constants relevant to the detailed band structure of the conduction electrons. In the Eq. (1), the first and the second terms represent the skew scattering and the side jump effect, respectively. In the present case, the temperature dependence of ρ, as shown in Fig. 3, is monotonic and smooth even around $T_f(H)$, 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_{xy} and the M. Together with this monotonic $\rho(T)$, the behavior of ρ_{xy}/M in Fig. 2 clearly indicates that one has to include another term in Eq. (1).

Tatara and Kawamura [10] 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 [10]:

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

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.* [19]. 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 [10]. They have shown that ρ_{xy}/M is expressed as follows:

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

where X_χ, X_χ^{nl} are constants with respect to M. The above argument contains two physically important meanings. First, ρ_{xy}/M no longer depends on M when M is sufficiently small. Because $\rho(T)$ is monotonic, the temperature dependence of observed ρ_{xy}/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 $H_{\rm so}$, by the uniform magnetization M means that M acts as a "symmetry-breaking field" of χ_0 [10,11]. Therefore, M is regarded as a "chiral field," and the constant X_χ, $X_\chi^{\rm nl}$ in Eq. (3) are a "chiral linear susceptibility" 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 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 [11], as exactly evidenced experimentally in Fig. 2.

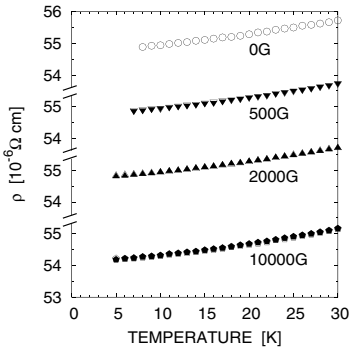

FIG. 3. Temperature dependence of resistivity ρ under ZFC and FC protocols in the fields indicated.

The *H-T* phase diagram deduced from the ρ_{xy} and the M measurement is shown in Fig. 4 together with the data obtained in Ref. [20]. The present *H-T* phase diagram is qualitatively consistent with those obtained from the torque measurement. The quantitative difference is due to the different definitions of $T_f(H)$. In the lower field ($H < 2000$ G), $T_f(H)$ behaves like AT and deviates from AT at the higher field region. This behavior has been interpreted as the crossover from AT to GT region.

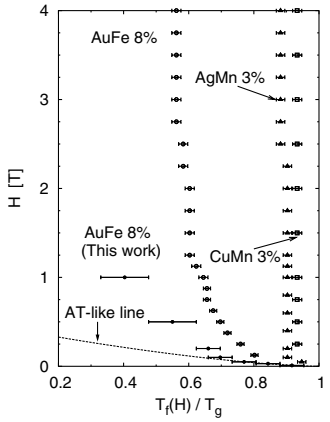

FIG. 4. *H-T* phase diagram. The data of CuMn 3%, AgMn 3%, and AuFe 8% (open circle) are taken from Ref. [20].

According to the mean field interpretation, spins cant below the GT transition temperature. The freezing of the transverse spin component certainly leads to the appearance of a local chirality. The chirality scenario of SG transition also predicts the similar crossover [21], although the mechanism of the SG transition in a field is quite different from that of the mean field theory.

The SG transition with the freezing of the transverse spin component is also expected in the reentrant SG. This transition was clearly observed by Mössbaur effect [22] in AuFe 19 at. % Fe. A cusplike anomaly of the Hall resistivity at reentrant SG transition temperature was recently observed in manganite reentrant SG [23] and in reentrant SG alloy FeAl [24]. For 18 at. % Fe, we made the same simultaneous ρ_{xy} and M measurements, where the anomalies in ρ and ρ_{xy} were observed at $T_c = 180$ K. Figure 5 shows the temperature dependence of ρ_{xy}/M for the AuFe 18 at. % Fe sample. One can see that the ZFC data has a significant decrease below 20 K which is identical to the reentrant SG transition temperature. In contrast, however, the absence of the derivation between the ZFC and FC curves has been observed for the ferromagnetic AuFe 28 at. % Fe sample. We believe that the

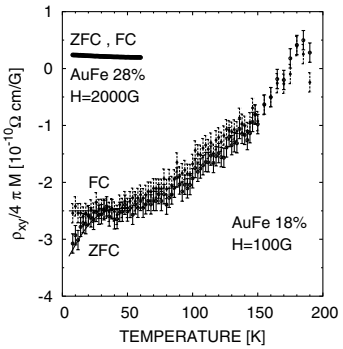

FIG. 5. Temperature dependence of ρ_{xy}/M for AuFe 18 at. % Fe (reentrant SG) and AuFe 28 at. % Fe (ferromagnet).

anomaly below 20 K is caused by the possible chirality contribution which associates with the reentrant SG.

In summary, we have carried out the simultaneous measurements of the extraordinary Hall resistivity and the magnetization of a canonical spin glass AuFe 8 at. % Fe. The temperature dependence of ρ_{xy}/M, which represents the uniform chiral susceptibility, shows a cusp at T_g and the ZFC and FC hysteresis below T_g. An anomaly was also observed in ρ_{xy}/M in ZFC below the reentrant SG transition temperature of the AuFe 18 at. % Fe system. These observations are compatible with the prediction by the Kawamura's chirality scenario of the canonical SG [11].

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