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Anomalous and spin Hall effects in magnetic granular films

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We propose a simple method to confirm the crossover effect; a change in the power-law dependence of the anomalous and spin Hall conductivities with respect to the longitudinal resistivity for magnetic granular films exhibiting giant magnetoresistance (GMR). Adopting a phenomenological theory, which explains the GMR in magnetic granular films, we show that a higher order term of the longitudinal resistivity appears in the Hall resistivity owing to the crossover effect. The result may resolve a controversy in the anomalous Hall effect observed in a magnetic granular film.

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The spin Hall effect (SHE) and anomalous Hall effect (AHE) with intrinsic origins have recently attracted much interest.[1,2] Following the discovery that electron scattering is crucial for both SHE and AHE,[3–5] theoretical studies have accounted for extrinsic and intrinsic origins and have uncovered several important results: (1) the spin Hall conductivity (SHC) in transition metals is much larger than that in semiconductors and is positive (negative) for late (early) transition metals.[6–8] (2) The orbital degree of freedom is important for the SHE in transition metals and their compounds.[9] (3) The SHC in the low-resistivity regime is governed by the Berry phase[10] and almost independent of the longitudinal resistivity ρ but shows a crossover with increasing ρ and decreases as $1/\rho^2$ at high resistivities.[6] These results have been obtained by using a realistic tight-binding (TB) model with atomic LS coupling. (4) As for the anomalous Hall conductivity (AHC), two crossovers have been predicted.[11,12] One occurs between the high-conductivity regime, where the AHE is dominated by skew scattering (SS),[13] and the ρ-independent regime, where the side jump (SJ) (Ref. 14) and Berry phase, that is, Karplus-Luttinger[10] mechanisms, are dominant. The other occurs between the ρ-independent regime and the high-resistivity regime. In the high-resistivity regime, the AHC decreases as $1/\rho^{\delta}$ with $\delta \sim 1.6$. The latter result has been obtained for a spin-polarized two-dimensional electron gas (2DEG) with a Rashba spin-orbit interaction and compared with many experimental measurements.

Because the SHE is a spin-current version of the AHE, the power law of the AHC and SHC in the high-resistivity regime should be the same. Therefore, the disagreement in the power law of the SHC and AHC predicted is an issue to be resolved. The ρ dependence of the AHC has been reexamined by Kovalev *et al.*[15] using the same model as that of Onoda *et al.*[11,12] They concluded that the power-law AHC $\sim 1/\rho^{1.6}$ appears only in a narrow range of ρ. The result by Kovalev *et al.*[15] is only applicable to low-ρ regime, and we should be cautious with the scaling relation in higher-ρ regime. Since the power law of the Hall conductivity is closely related to the mechanism of the Hall effects, it is an emergent issue to investigate the power law experimentally.

The AHE is usually analyzed in terms of a simple relation between the Hall resistivity ρ_H and ρ, namely, $\rho_H = a\rho + b\rho^2$, where the first and second terms come from the SS and SJ mechanisms, respectively. This relation is derived from the fact that the AHC depends on $1/\rho$ for SS, while it is independent of ρ for SJ. When the AHC varies as $1/\rho^{\delta}$, a term $c\rho^{2+\delta}$ may also appear in ρ_H. Therefore, careful measurements of ρ_H at high resistivities may make it possible to identify the power law. The analysis, however, should be done on the same kind of samples to resolve the controversy. Recent experiments on the AHE for doped $SrTiO_3$ have suggested that δ can be 2.[16] In this Rapid Communication, we propose that a magnetic granular film (MGF) (Refs. 17 and 18) is an ideal system to study since the resistivity of MGFs is relatively high and varies with the magnetic field H, leading to giant magnetoresistance (GMR). Indeed, over a decade ago, Xiong *et al.*[19] reported an anomalous power law $\rho_H \propto \rho^n$ with $n = 3.7 \pm 0.2$ for Co-Ag MGFs, the origin of which remains unknown.

In this Rapid Communication, we propose that the AHE in MGFs is closely related to the SHE in the nonmagnetic host metal once the resistivity becomes spin dependent. Therefore, it makes sense to first calculate the SHC in the noble metals Ag and Au to confirm the crossover and deduce a simple formula for the relation between ρ_H and ρ using the spin-dependent resistivity (two-current model)[20,21] that explains the GMR effect in MGFs and magnetic multilayers. It will be shown that the expression for ρ_H includes higher order terms such as $\rho^{2+\delta}$. The formula will be used to reexamine the data obtained by Xiong *et al.*[19] to show that $\delta \sim 1$. This result will be discussed from both theoretical and experimental viewpoints.

MGFs are composed of ferromagnetic grains of a few nm radius embedded in a nonmagnetic host material such as Cu, Ag, or Au. Without an external magnetic field, the direction of the magnetic moments of the grains is random and no magnetization appears. Application of a large magnetic field makes the magnetic moments of the grains align so that the resistivity ρ drops (GMR effect) and an AHE appears. The effect of GMR has been explained in terms of spin-dependent scattering of conduction electrons at the interface of the magnetic grains.

We briefly review the phenomenological theory of GMR.

Let $\rho_{+(-)}$ be the resistivity of the majority (minority) spin electrons scattered at the interface of a grain. When the magnetic moment of the grain makes an angle θ with the external magnetic field $\boldsymbol{H}$, the up (down) spin resistivity in the global spin axis parallel to $\boldsymbol{H}$ is

$$\rho_{\uparrow(\downarrow)}(H) = \{\rho_+ + \rho_- + (-)\cos\,\theta(\rho_+ - \rho_-)\}/2. \quad (1)$$

Since the direction of the local magnetic moments of the grains is random, we average $\cos\,\theta$ over the distribution of θ and denote it as $\langle\cos\,\theta\rangle$. The dependence of $\langle\cos\,\theta\rangle$ on H may be complicated in general, but for simplicity we assume that $\langle\cos\,\theta\rangle \sim M(H)/M_S \equiv m$, where M_S is the saturation magnetization. The total resistivity is

$$\rho(H) = \frac{1}{4}\left[\rho_+ + \rho_- - m^2\frac{(\rho_+ - \rho_-)^2}{\rho_+ + \rho_-}\right]. \quad (2)$$

The MR ratio is defined as

$$MR(H) = \frac{\rho(0) - \rho(H)}{\rho(H)}. \quad (3)$$

We find that the larger the difference $\rho_+ - \rho_-$ is, the larger the MR ratio is. The GMR effect of MGFs has been explained by numerical simulations based on a spin-dependent resistivity model. The m^2 dependence of $\rho(H)$ has been confirmed in both experiments and numerical simulations.[18,21]

The AHE in MGFs may be explained in the following way. Under an external electric field, a Hall current is produced within each magnetic grain but decays in the nonmagnetic host material when $H=0$ and no AHE occurs. However, the Hall current may be finite when the magnetic field H aligns the magnetic moments of the grains and makes the host material spin polarized. The origin of this type of AHE may be attributed to the usual SS or SJ mechanisms.

In this work, we put forward another explanation of the AHE of MGFs in terms of the spin-dependent resistivity. When $H=0$, the direction of the magnetic moments of the grains is random and the system is paramagnetic. Although the resistivity is then independent of spin ($\rho_\uparrow = \rho_\downarrow$), a SHE may exist.[22] When $H \neq 0$, then $\rho_\uparrow \neq \rho_\downarrow$, and the resistivity dependence of the SHC results in a difference between $\uparrow$ and $\downarrow$ spin Hall conductivity, which is the anomalous Hall conductivity. The AHC may be estimated from the energy dependence of the SHC calculated for the nonmagnetic material. The AHC thus estimated agrees quite well with that calculated directly for the spin-polarized state.

Now, we calculate the SHC as a function of the resistivity for Ag and Au adopting a full-orbital tight-binding model with the atomic LS coupling. The electronic structures were calculated using the NRL-TB model by taking into account the hopping parameters up to sixth neighbor sites.[8] Figure 1 shows the electronic structure calculated for Ag, where zero energy is the Fermi energy. The dotted line labeled $n=10$ corresponds to the Fermi energy of Pd.

The SHC is calculated using the Kubo-Streda formula with lifetime broadening γ (imaginary part of the self-energy) caused by electron scattering due to the randomness. The current vertex corrections were found to be small and were therefore neglected. The resulting SHCs for Ag and Au

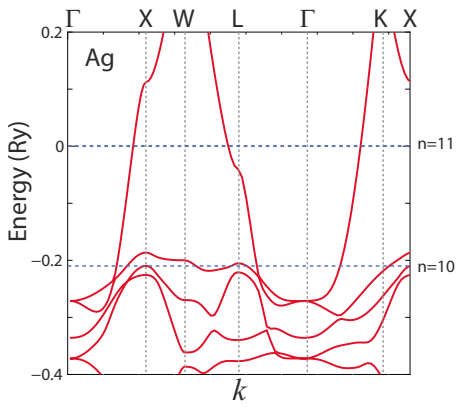

FIG. 1. (Color online) Electronic structure of Ag calculated using the full-orbital TB model with atomic LS coupling.

are plotted in Fig. 2 on a logarithmic scale. The right-hand axis is the resistivity estimated from γ values. In the calculation of the SHC, we tried two approximations: constant γ for the self-energy and the Born approximation. The results indicated that the constant γ approximation is more appropriate. The SHC is nearly constant below $\rho=10\ \mu\Omega$ cm and the crossover occurs around $\rho=10\sim100\ \mu\Omega$ cm. The same tendency occurs for the SHC of Pt.[6] The results at low resistivity are consistent with those calculated from first principles.[23]

Since the resistivity of MGFs is rather high, namely, $\rho=5\sim20\ \mu\Omega$ cm, both origins (from magnetic grains and from spin-dependent resistivity) mentioned above should be taken into account for the AHE in MGFs. The distinction of these origins, however, is not clear enough since the spin-dependent resistivity may be caused by scattering at the interfaces and inside the grains as well. We therefore use a phenomenological theory to incorporate both mechanisms to explain the dependence of ρ_H on ρ.

The AHC due to the SS and SJ effects may arise from a

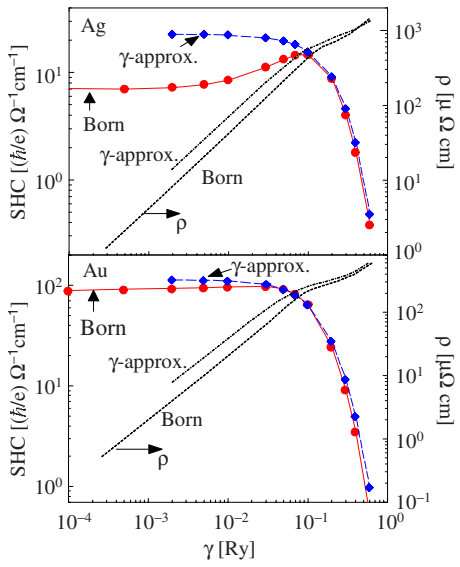

FIG. 2. (Color online) Calculated SHC as a function of lifetime broadening γ for Ag (upper panel) and Au (lower panel) on a logarithmic scale.

difference in the charge Hall conductivity of up (↑) and down (↓) spin states as Crepieux and Bruno[24] showed in a microscopic theory. The crossover in the SHC shown in Fig. 2 should also be present in the AHC of magnetized MGFs. The crossover effect can be incorporated with a contribution from the SJ mechanism in the AHC since the SHC is caused by the anomalous velocity as AHC in the SJ mechanism. Thus the following expressions are adopted for the AHC from SS and SJ contributions using the spin-dependent resistivity $\rho_{\uparrow(\downarrow)}$,

$$\sigma_{xy}^{SS}(H) = \frac{a_{\uparrow}(H)}{\rho_{\uparrow}(H)} - \frac{a_{\downarrow}(H)}{\rho_{\downarrow}(H)}, \tag{4}$$

$$\sigma_{xy}^{SJ}(H) = \frac{b_{\uparrow}(H)}{1+(\rho_{\uparrow}(H)/\rho_{*\uparrow})^{\delta}} - \frac{b_{\downarrow}(H)}{1+(\rho_{\downarrow}(H)/\rho_{*\downarrow})^{\delta}}, \tag{5}$$

where $a_{\uparrow(\downarrow)}(H)$ and $b_{\uparrow(\downarrow)}(H)$ are coefficients, which depend on the magnetic field in general, and $\rho_{*\uparrow(\downarrow)}$ is the crossover resistivity. Since $\rho_{\uparrow(\downarrow)}(H) \ll \rho_{*\uparrow(\downarrow)}$ in MGFs, the right-hand side of Eq. (5) can be expanded as

$$\sigma_{xy}^{SJ}(H) \sim b(H) - \left(\frac{b_0}{\rho_*^{\delta}}\right)(\rho_{\uparrow}^{\delta}(H) - \rho_{\downarrow}^{\delta}(H)), \tag{6}$$

where $b(H)=b_{\uparrow}(H)-b_{\downarrow}(H)$, by assuming $b_0 \equiv b_{\uparrow}(H) \sim b_{\downarrow}(H)$ and $\rho_* \equiv \rho_{*\uparrow} \sim \rho_{*\downarrow}$ in the second term for simplicity. Since the resistivity in MGFs is usually high and the contribution from SS is weak, we also assume that $a_{\uparrow}(H) \sim a_{\downarrow}(H) \equiv a$.

By using expressions for $\rho_{\uparrow(\downarrow)}$, the relation $\rho_H(H) = \rho^2(H)(\sigma_{xy}^{SS}(H)+\sigma_{xy}^{SJ}(H))$ results in

$$\rho_H(H) = a\ \mathrm{sgn}(\rho_- - \rho_+)\sqrt{MR}\rho(H)m + b(H)\rho^2(H) - \left(\frac{4b_0}{\rho_*^{\delta}}\right)\rho^{\delta}(0)\rho^2(H)[\{1+\mathrm{sgn}(\rho_+ - \rho_-)m\sqrt{MR}\}^{\delta} - \{1-\mathrm{sgn}(\rho_+ - \rho_-)m\sqrt{MR}\}^{\delta}]. \tag{7}$$

The first and second terms correspond to the usual SS and SJ contributions, respectively, and the last term is caused by the crossover effect.

Using Eq. (7), the experimental results for MGFs may be analyzed. Since the resistivity is relatively high, the contribution from SS may be neglected. In the paramagnetic state for $H=0$, it follows that $m=0$ and $b=0$, and therefore the Hall resistivity is zero. The longitudinal resistivity in this state is referred to as ρ_{AP}. With increasing H, the magnetization becomes finite and the resistivity drops. When the magnetization is saturated, $m=1$ and $\rho(H)$ tends to a constant value ρ_P. Consequently, Eq. (7) becomes

$$\rho_H = b(H)\rho_P^2 + \mathrm{sgn}(\rho_- - \rho_+)\left(\frac{8b_0}{\rho_*^{\delta}}\right)\rho_{AP}^{\delta}\rho_P^2\sqrt{MR}, \tag{8}$$

with $\delta=1$ or 2. This is the main result of our analysis. The first term comes from the usual SJ contribution of the ferromagnetic grains, and the second term is caused by the crossover effect. When the difference between ρ_P and ρ_{AP} is not large, then $\rho_P \sim \rho_{AP} \equiv \rho$, and ρ_H contains a higher order term $\rho^{2+\delta}$.

Now consider the exponent δ in Eq. (8) simplified as $\rho_H = b_1\rho_P^2 + b_2\rho_{AP}^{\delta}\rho_P^2\sqrt{MR}$. Xiong *et al.*[19] measured the GMR and AHE in Co-Ag MGFs by changing the grain size, achieved by changing the annealing temperature of the samples. The idealized experimental results for $\rho(H)$ and ρ_H are plotted in the insets of Fig. 3. By using the values of ρ_P, ρ_{AP}, and the MR ratio, ρ_H/ρ_P^2 is plotted as a function of $\rho_{AP}^{\delta}\sqrt{MR}$ for $\delta=1$ and 2 in Fig. 3. Linearity is achieved for $\delta=1$, suggesting that a higher order contribution to ρ_H is ρ^3. The exponent, however, agrees neither with $\delta=2$ nor $\delta=1.6$ predicted theoretically.

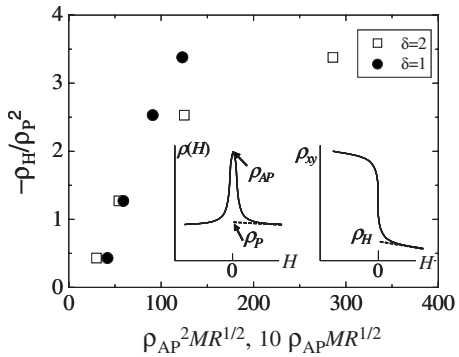

FIG. 3. Analysis using Eq. (8) and experimental results. The left inset shows a schematic dependence of the resistivity on H, and the right one shows the corresponding ρ_{xy}. Definitions of ρ_P, ρ_{AP}, and ρ_H used in the analysis are also indicated in the insets.

Several reasons may explain the disagreement between the present analysis and the theoretical predictions. The range of the resistivity observed is between 5 and 20 $\mu\Omega$ cm, which is too narrow to fully represent the crossover, and the resistivity observed may be much smaller than $\rho_* \sim 10^2$ $\mu\Omega$ cm. Variation in the SHC by using Ag-Pd and Au-Pt alloys, for example, might give additional information since the SHC of these alloys increases with decreasing Fermi energy as shown in Fig. 4.

The sign of the Hall resistivity is consistent with the experimental results. The sign change observed is also reproduced in the present analysis.[25] The higher order power law for ρ_H with respect to ρ may appear also in the AHE for magnetic multilayers.[26–28] The contribution from the crossover effect, however, may be weak because the resistivity in magnetic multilayers is usually smaller than that in MGFs. Nevertheless, inclusion of the crossover effect in the micro-

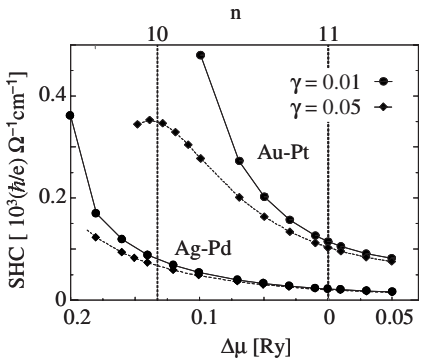

FIG. 4. SHC for Ag-Pd and Au-Pt alloys calculated from the rigid-band model for the lifetime broadening $\gamma=0.01$ and 0.05.

scopic analysis for magnetic multilayers[29,30] would be useful.

We have neglected effects of spin-flip scattering in the analysis. The spin-flip scattering may increase in MGFs with low annealing temperature in which isolated magnetic impurities remain. In these samples the MR effect would not saturate with increasing H, and the simple two-current model might fail to explain the GMR. In our analysis, we have applied Eq. (8) to MGFs in which the GMR effect is clearly identified as in magnetic multilayers. We expect that the spin-mixing conductance in such MGFs may be the same order of magnitude with that in magnetic multilayers[31] and will not alter the qualitative result at least the exponent in the $\rho_H - \rho$ relation.

In conclusion, we have calculated the SHC of Ag and Au in the full-orbital TB model with LS coupling and showed that a crossover appears. Adopting a phenomenological model including the crossover effect, we obtained a simple relation between the Hall resistivity ρ_H and the longitudinal resistivity ρ and analyzed the AHE in Ag-based MGFs, which show the GMR effect. The present analysis shows that a higher order term of ρ exists in ρ_H observed for Ag-Co MGFs. However, the power law is not consistent with the theoretical predictions. More detailed experiments for high-resistivity regime are desirable to obtain a conclusive result on the power law.

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