Evolution of Anomalous Hall Behavior in Thin Pt/Co/Pt Trilayers

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Abstract

In this work, through controlling spin scattering mechanisms, anomalous Hall behaviors exhibit a series of evolutions in thin Pt/Co/Pt trilayers. The shape of Hall resistivity over longitudinal resistivity (ρ_{AH}/ρ_{xx} versus ρ_{xx}) curve turns from bending to linear and then bending again in most trilayers. This kind of evolution cannot be explained by the conventional linear scaling of anomalous Hall effect. It should be ascribed to the contribution of spin-phonon skew scattering. Our research may help to understand spin scattering behavior in low-dimensional systems more deeply and build a proper synergy between theory and experiment on the research of anomalous Hall effect.

Keywords:

Pt/Co/Pt, trilayer, anomalous Hall effect, scaling

1. Introduction

Some recent hot research topics such as spin Hall effect[1], spin Seebeck effect [2], magnetic proximity effect [3, 4], anomalous Nernst effect [5, 6, 7] and spin torques[8, 9] have been found being closely involved with spin orbit coupling (SOC). As the earliest discovered magneto-transport phenomena, anomalous

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Hall effect (AHE) attracts interest once again because it is unambiguously one of the most powerful methods to explore the SOC. Also, AHE has potential application in high sensitivity Hall sensors [10, 11, 12]. In general, anomalous Hall resistivity (ρ_{AH}) is determined by the skew scattering ($\propto \rho_{xx}$) [13, 14], intrinsic Berry curvature of energy bands [15] ($\propto \rho_{xx}^2$), and side-jump mechanism [16] ($\propto \rho_{xx}^2$), where ρ_{xx} is longitudinal resistivity. Then a conventional expression can be phenomenologically written as Eq. (1):

$$\rho_{AH} = a\rho_{xx} + b\rho_{xx}^2. \quad (1)$$

If the material, for example Mn_5Ge_3 [17], has a strong temperature dependent spontaneous magnetization (M), the parameters a and b must be modified according to the temperature dependent function $f(T)$, and thus Eq. (1) was revised as

$$\rho_{AH} = af(T)\rho_{xx} + bf(T)\rho_{xx}^2. \quad (2)$$

Although there had been lots of debates on the origins of AHE in theory, explanations for experimental works were usually based on the above two equations for a long time until 2009. In this year, Tian Y. et al. [18] pointed out that the skew scattering item should be divided into two parts according to Matthiessen rule: one stems from impurity scattering and the other from electron-phonon scattering. The phenomenological expression should be modified by a new scaling,

$$\rho_{AH} = a'\rho_{xx0} + a''\rho_{xxT} + b\rho_{xx}^2, \quad (3)$$

where ρ_{xx0} is residual resistivity and ρ_{xxT} is resulted from thermal activity. Furthermore, some researchers tried to refine the new scaling and introduced more parameters, which weakens the creditability of the new scaling somehow [19, 20].

Up to now, the validity of the new scaling rule is not fully explored, while the conventional scaling is being frequently used. The conventional scaling displays a linear behavior of Hall angle (i.e. ρ_{AH}/ρ_{xx}) versus ρ_{xx} . However, it is obvious that the new scaling exhibits a nonlinear dependence of Hall angle on ρ_{xx} (see Eq. (3)) since there is no reason to always make $a' = a''$. Thus a natural

question was raised: why such a linear relationship between Hall angle and longitudinal resistivity can be used to explain previous observations if the new scaling (nonlinear) is valid?

To answer and clarify all these questions, we therewithal prepared a series of Pt/Co/Pt trilayers with an equal number of spin scattering sources, in which the competition among the three mechanisms could be changed by post-annealing temperatures (T_a).

2. Experiments

Pt (t nm)/Co (0.5 nm)/Pt (5- t nm) trilayers with thickness (t) of 1, 1.75, 2.5, 3.25 and 4 nm were prepared on MgO(100) substrates by direct current sputtering under the Ar pressure of 0.52 Pa (Pt) and 0.35 Pa (Co). The base pressure of sputtering chamber was better than 2×10^{-6} Pa. All samples were covered by the Hall bar mask during sputtering for Hall measurement. Magnetic properties of the samples were characterized by a vibrating sample magnetometer (VSM, LakeShore Inc.) and a VersaLab system (Quantum Design, Inc.). AHE measurements were carried out from 20 to 300 K, with each sample being measured both as-deposited and after post-annealing at 100, 200, 300, 400 and 500 °C for one hour.

3. Results and Conclusions

Empirically, the quality of the ferromagnetic layer would be improved after post-annealing below a critical temperature, and then takes a turn for the worse with higher annealing temperatures because of interfacial diffusion. The critical annealing temperature depends on the substrate, buffer layer, cap layer and the ferromagnetic layer itself. Figure. 1 shows a diagrammatic sketch of Co atom distribution in Pt/Co/Pt trilayers under different thermal treatments: as-deposited (a), post-annealed at low temperatures (b) and post-annealed at high temperatures (c). Hysteresis loops of Pt (2.5 nm)/Co (0.5 nm)/Pt (2.5 nm) trilayers with various annealing temperatures are shown in Fig. 1(d) as an

example. It is well known that the perpendicular magnetic anisotropy (PMA) originates from the interfacial interaction in Pt/Co/Pt trilayer [21, 22, 23]. A better interface will apparently enhance the interfacial orbital hybridizations and contribute to an out-of-plane coercivity enhancement. Corresponding to Fig. 1(d), the out-of-plane coercivity reaches a maximum at T_a around 200 °C, and then decreases. PMA dies away due to gradually destroyed Pt/Co interface after overheating. By the way, the critical T_a is 200 °C for Pt (3.25 nm)/Co (0.5 nm)/Pt (1.75 nm) and Pt (4 nm)/Co (0.5 nm)/Pt (1 nm) trilayers also, while it is around 300 °C for another two trilayers. During this process, inevitably, skew scattering, side-jump and intrinsic Hall conductivity are changing with an invariant number of magnetic atoms in Pt/Co/Pt trilayer. Meanwhile, the influence of bypassing current through Pt layer would be the same for all trilayers. The competition among them must be reflected on AHE according to Eq. (1) or (3). Moreover, skew scattering can be enhanced by the oxide layer and the vacuum interface [24, 25].

Dimensions of the Hall bar samples, temperature dependences of longitudinal resistivity and partial anomalous Hall resistivity loops for Pt (2.5 nm)/Co (0.5 nm)/Pt (2.5 nm) trilayer are shown in Fig. 2. The shift of longitudinal resistivity curves are not significant after post-annealing below 400 °C, as shown in Fig. 2(b). The fluctuation of longitudinal resistivity curve for the as-deposited sample (black line in Fig. 2(b)) at low temperature might be ascribed to substrate roughness or imperfect interface quality of the as-deposited sample. The trends of all curves are almost the same. Because of the same reason shown in Fig. 1, the longitudinal resistivity becomes smaller when the trilayer was post-annealed below 200 °C and then increases after annealing at higher T_a . Due to the bad interface caused by annealing at 500 °C, the resistivity became dramatically large and Hall measurement was failed. Fig. 2(c) exhibits the anomalous Hall resistivity loops for Pt (2.5 nm)/Co (0.5 nm)/Pt (2.5 nm) trilayer measured at 300 and 20 K.

Detailed relationships between ρ_{AH}/ρ_{xx} and ρ_{xx} are shown in Fig. 3. As we analyzed above, skew scattering, side-jump and intrinsic contributions are

Figure 1: (Color online) Diagrammatic sketch of Co atom distribution in Pt/Co/Pt trilayers under different thermal treatments: (a) as-deposited, (b) post-annealed at low temperatures, (c) post-annealed at high temperatures. (d) hysteresis loops of Pt (2.5 nm)/Co (0.5 nm)/Pt (2.5 nm) trilayers with various annealing temperatures.

Figure 2: (Color online) Profile of anomalous Hall effect measurement for Pt (2.5 nm)/Co (0.5 nm)/Pt (2.5 nm) trilayer. (a) Diagrammatic sketch of the Hall bar, (b) temperature dependence of longitudinal resistivity, (c) anomalous Hall resistivity loops at 300 and 20 K. T_m is measurement temperature.

Figure 3: (Color online) ρ_{AH}/ρ_{xx} versus ρ_{xx} curves of the trilayers post-annealed at different temperatures. Solid lines are fitted by $\rho_{AH} = a'\rho_{xx0} + a''\rho_{xxT} + b\rho_{xx}^2$.

changed by the thermal treatment in all five samples. The varying competition among these origins of AHE directly participates in the shape evolution of Hall angle curves. Except for the trilayer Pt (1 nm)/Co (0.5 nm)/Pt (4 nm), the shape of Hall angle curves alters from bending (RT, 100 °C) to nearly linear (200 °C) and then becomes bending (300 °C, 400 °C) again. This kind of behaviors obviously cannot be explained by the conventional scaling expressed by Eq. (1). Temperature dependence of spontaneous magnetization is weak for Pt/Co/Pt trilayer, which we do not give here because of a large noise amplitude in this measurement. Less than 10% variation of the spontaneous magnetization from 20 to 300 K could perhaps be neglected according to the results in Pt/Co/Pt multilayers [20]. Also, the temperature dependence of spontaneous magnetization is monotonic for Pt/Co/Pt trilayer. Here $f(T)$ revision is not enough for the fitting either. For a comparison, all data can be fitted well by the new scaling, as shown by the solid lines in Fig. 3.

To clarify the relationship between the shape of Hall angle curves and the parameters in the new scaling, correlation coefficient (R) [26] (a measure of the linear correlation between two quantities) is introduced. It is defined as the covariance of variables X and Y divided by a product of their standard deviations,

$$R = \frac{\text{Cov}(X, Y)}{\sigma_x \sigma_y} = \frac{\sum_{i=1}^N (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^N (x_i - \bar{x})^2 \sum_{i=1}^N (y_i - \bar{y})^2}}. \quad (4)$$

where N is the number of data points, $\text{Cov}(X, Y)$ is the covariance [26], $\sigma_{x(y)}$ is the standard deviation of variable $X(Y)$, x_i represents the experimental data of longitudinal resistivity. y_i is the Hall angle. For a specific i , y_i is generated in such a way that ρ_{xx} and ρ_{xx0} are taken from experiment for i -th datum, while a''/a' and $b\rho_{xx0}/a'$ serve as independent variables (i.e. x and y axes of 3D R map). Mathematically, any one of a' , a'' and $b\rho_{xx0}$ can be the basic unit for R map. We chose a' and plotted R map against a'' and $b\rho_{xx0}$ only for convenience. $\bar{x}$ and $\bar{y}$ are averaged values of ρ_{xx} and ρ_{AH}/ρ_{xx} . The small balls indicate a series of R values under the coordinates of $(a'', b\rho_{xx0})$. Here, $R = 1$ means that the linear relationship of ρ_{AH}/ρ_{xx} versus ρ_{xx} has a positive slope, while $R = -1$

Figure 4: (Color online) 3D maps of correlation coefficients (R) of ρ_{AH}/ρ_{xx} and ρ_{xx} versus a'' and bp_{xx0} in the unit of a' for Pt (t nm)/Co (0.5 nm)/Pt (5- t nm) trilayers with thickness (t) of 1, 1.75, 2.5, 3.25 and 4 nm. Here, a' and a'' indicate the contributions of skew scattering from impurities and phonons; b represents the intrinsic and side-jump contributions; ρ_{xx0} is the resistivity of the trilayer at 20 K.

means a negative one. $-1 < R < 1$ means a nonlinear curve shape.

It is obvious that all the samples annealed at different temperatures obey the same R distribution, which is similar to a cliff-like shape. The shape evolution of the Hall angle curves with annealing temperatures can be directly displayed by the trajectory of R values of annealed samples (spherical ball) on the R map. Hall angle curves are nearly linear in vast configurations of a' , a'' and b , even though the new scaling does not seem to exhibit a linear relationship between ρ_{AH}/ρ_{xx} and ρ_{xx} . Only for the configuration located in a limited area ($-1 < R < 1$), Hall angle curve is in bending shape. These 3D maps mathematically reveal the reason why linear behaviors of Hall angle curves were usually observed in reported researches on AHE, especially in the case of $b\rho_{xx0}/a' > 0$, as shown in Fig. 4. When Co layer is close to MgO substrate or vacuum, interfacial scattering is largely enhanced and thus their evolution paths are similar in Fig. 4(a) and (e) but different from those in (b), (c) and (d). Besides, annealing temperature dependence of R value shown in Fig. 4(f) greatly coincides with the dependence of coercivity and longitudinal resistivity of the trilayers based on the same reason we stated before.

The reliability of fitting results on AHE is another challenging issue uncovered by Fig. 4. For the case of bending curves of Hall angle ($-1 < R < 1$), the nonlinear fitting by the new scaling is credible on the whole since the corresponding R value distributes in a limited small region (the cliff region of the 3D R map). The linear region corresponds to multiple solutions when the measurement error is larger than the fitting error, which is hardly avoided experimentally. For example, Hall angle curve of Pt (3.25 nm)/Co (0.5 nm)/Pt (1.75 nm) trilayer annealed at 200 °C can be fitted well by both scalings, as shown in Fig. 5(a), but the fitted parameters are totally different. More calculations proved that if $b\rho_{xx0}$ is two orders larger than a' , the Hall angle curves are linear and almost overlapping in a wide range of a'' , as shown in Fig. 5(b).

For bulk materials and thick films, $b\rho_{xx0}$ is eminently larger than a' , so the conventional scaling can be used in lots of AHE works. For AHE studies on thinner films with suppressed Hall conductivity owing to the dimension effect,

Figure 5: (Color online) Reliability demonstration of fitting results for the sample with a linear trend of ρ_{AH}/ρ_{xx} versus ρ_{xx} . (a) Experimental data of ρ_{AH}/ρ_{xx} versus ρ_{xx} (scattered) and fitting curves (solid) for Pt (3.25 nm)/Co (0.5 nm)/Pt (1.75 nm) trilayer annealed at 200 °C. (b) Calculated ρ_{AH}/ρ_{xx} versus ρ_{xx} curves by using ρ_{xx} in (a) and given parameters of a' , a'' and b .

the fitted data should be carefully disposed.

Based on the above analysis, a qualitative explanation is proposed to establish the physical picture of AHE when changing the dimension of the system and the annealing process. Inspired by the results of Fig. 5(b), the competition between skew scattering and side-jump may determine the shape of Hall angle curve. It is noted that the Hall angle versus ρ_{xx} is almost linear when the side-jump mechanism is dominant (b term is much larger). When the side-jump scattering is weakened to be comparable to the skew scattering (a term), the cooperation of these two contributions leads to a bending curve of Hall angle with ρ_{xx} . Referring to Berger's theory [9], the side-jump contribution depends on an enhancement factor τ_q which is related to the periodic lattice structure. For ultrathin ferromagnetic film (0.5 nm Co film in this work), the periodic lattice structure is easier to be distorted due to less neighboring atoms, surface roughness and annealing temperature. Therefore, the side-jump contribution ($b\rho_{xx}$) may be expected to be reduced but skew scattering is enhanced in the presence of non-negligible interfacial scattering [24, 25], so that the nonlinear curve of Hall angle versus ρ_{xx} may be induced. On the other hand, for thick films or bulk materials, a linear relationship between Hall angle and ρ_{xx} occurs because of the increased τ_q . In our case, since the thickness of cobalt layer (0.5 nm) is thin enough, most of ρ_{AH}/ρ_{xx} versus ρ_{xx} curves are bending. Similar phenomenon for thin ferromagnetic film can be observed in Ref. [18] although this bending is weak there. Moreover, it is observed in Fig. 3 that the Hall angle curves become straight lines because a better quality of crystal can be formed at the optimal 200°C post-annealing temperature. In this case, the enhancement factor for the side-jump contribution becomes larger, giving rise a linear line. At other annealing temperatures, the perfect periodic lattice of the Co film will be deviated so that the enhancement factor of side-jump will be reduced, and bending curves are then derived in accordance to this reduction.

4. Conclusions

In summary, shape evolution of Hall angle curves was demonstrated in Pt/Co/Pt trilayers, which supports the new scaling of AHE. 3D maps of correlation coefficient for ρ_{AH}/ρ_{xx} and ρ_{xx} reveal a special distribution of the shape of Hall angle curves theoretically. The competition between side-jump and skew scattering may be the origin of the shape evolution of Hall angle curves. Our study may help to recognize the connection between theory and experiment on AHE in low-dimensional systems.

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