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Anomalous Hall effect and magnetoresistance behavior in Co/Pd_{1-x}Ag_x multilayers

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In this paper, we report anomalous Hall effect (AHE) correlated with the magnetoresistance behavior in [Co/Pd_{1-x}Ag_x]_n multilayers. For the multilayers with $n=6$, the increase in Ag content from $x=0$ to 0.52 induces the change in AHE sign from negative surface scattering-dominated AHE to positive interface scattering-dominated AHE, which is accompanied with the transition from anisotropy magnetoresistance (AMR) dominated transport to giant magnetoresistance (GMR) dominated transport. For $n=80$, scaling analysis with $R_s \propto \rho_{xx}^\gamma$ yields $\gamma \sim 3.44$ for $x=0.52$ which presents GMR-type transport, in contrast to $\gamma \sim 5.7$ for $x=0$ which presents AMR-type transport.

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Anomalous Hall effect (AHE) in magnetic materials was discovered more than a century ago.¹ Recently, the intensive interest in AHE has been promoted by its spin-dependent transport properties and technical applications.²⁻⁴ The Hall resistivity (ρ_H) was found to follow the form of the phenomenological equation

$$\rho_H = R_o B + 4\pi R_s M, \quad (1)$$

where the first term is the ordinary Hall resistivity due to the Lorentz force and the second term is anomalous Hall resistivity (ρ_{AH}), which is proportional to the out-of-plane magnetization of the film.⁴ To identify the origins of AHE, the scaling correlation between anomalous Hall coefficient R_s in Eq. (1) and zero-field longitudinal resistivity ρ_{xx} given by

$$R_s \propto \rho_{xx}^\gamma \quad (2)$$

is usually employed.² Skew scattering mechanism leads to the exponent $\gamma=1$,⁵ while both scattering dependent-side jump and scattering independent-intrinsic mechanism yield $\gamma=2$.⁶⁻⁸ For homogeneous ferromagnets, experimental γ values are in the range of 1–2, demonstrating the general coexistence of these mechanisms.^{9,10} However, for heterogeneous ferromagnetic systems, such as magnetic multilayers and granular films, interface scattering-dominated AHE has been proven to result in a large γ , for example, $\gamma=2.6$ in Fe/Cr multilayers,¹¹ $\gamma=2.6$ and 5.7 in Co/Pd multilayers,^{12,13} and $\gamma=3.7$ in granular Co-Ag films,¹⁴ while the mechanism of interface scattering on the value of γ is still missing in theory. On the other hand, in single layer films, such as Ni, surface scattering has been found to play a dominant role on AHE for film thickness less than 50 nm.^{15,16} In ferromagnetic/non-magnetic (FM/NM) multilayers, the sign of surface scattering-induced AHE has been found to be opposite to that of

interface scattering in Co/Pd multilayers, and the amplitude of the former could even overwhelm the latter.^{13,17}

In this paper, we studied AHE and magnetoresistance (MR) behavior in [Co(3 Å)/Pd_{1-x}Ag_x(5 Å)]_n multilayers with perpendicular magnetic anisotropy. With the increase of the substitution of Ag, the sign change in AHE accompanied with the increase of giant magnetoresistance (GMR) has been observed in the multilayers with $n=6$, which should be attributed to the transition from surface scattering-dominated AHE to interface scattering-dominated AHE.

Two series of Co/Pd_{1-x}Ag_x multilayer with a structure of Pd_{1-x}Ag_x(50 Å)/[Co(3 Å)/Pd_{1-x}Ag_x(5 Å)]_n were deposited by using magnetron sputtering on oxidized Si (100) substrates, with the Co/PdAg bilayers' repetition n of 6 and 80. A 10-nm-thick SiO₂ was deposited to cover the films to prevent from oxidation. The Pd_{1-x}Ag_x layers were deposited by co-sputtering, where the target power for Pd was fixed, by the means of varying the target power of Ag to adjust the Ag content. The Ag contents of $x=0, 0.04, 0.19, 0.32$, and 0.52 were obtained, which were measured by energy-dispersive spectrometer on relatively thick Co_{1-x}Ag_x films, larger than 200 nm. Hall loops were measured under magnetic fields up to 5 T applied perpendicular to the film plane. Detailed Hall bar fabrication and measurement can be found in Ref. 13.

Bogomolov *et al.* have studied the magnetic properties and structures of Co(4 Å)/Pd_{1-x}Ag_x(10 Å) multilayers.¹⁸ A significant increase in coercivity (H_c) from 1.4 kOe in $x=0$ to 2.7 kOe in $x=0.04$ has been observed. The origin of the increase in H_c by substituting Pd with Ag has been attributed to the increase of tensile stress because of the larger lattice constant of face-centered cubic (fcc) Ag (~ 4.086 Å) than that of fcc Pd (~ 3.883 Å). Further increase of the substitution results in a decrease in H_c , due to the decrease in the multilayer lattice matching, forming interface misfit dislocations.¹⁸

In this paper, we focused on magnetotransport properties in terms of AHE and MR in Co/Pd_{1-x}Ag_x multilayers. Shown in Fig. 1 are the representative Hall loops of [Co/Pd_{1-x}Ag_x]₆ multilayer with $x=0, 0.04, 0.19$, and 0.32

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measured at 5 K. All the samples present perpendicular magnetic anisotropy as square hysteresis loops could be obtained after removal of the linear background of ordinary Hall resistivity. H_c increases from 5.4 kOe for $x=0$ to 10.7 kOe for $x=0.04$. Considerably larger H_c in our samples than that of the samples of Bogomolov *et al.* should be attributed to the thinner Co layers in our samples, which demonstrates the nature of interface-induced perpendicular magnetic anisotropy in this structure.^{19,20} H_c decreases as x increases to more than 0.19. The dependence of H_c on x is in good agreement with that observed by Bogomolov *et al.*¹⁸

The AHE resistivity ρ_{AH} can be extracted from the Hall loops by extrapolating the high-field data from positive field to zero field, as shown in Fig. 1. The sign of AHE is negative for $x \leq 0.19$, while it is positive for $x \geq 0.32$, indicating the change in AHE sign with the substitution of Pd with Ag. Later analysis indicates the sign change from negative to positive is due to the transition from negative surface scattering-dominated AHE to positive interface scattering-dominated AHE.

The temperature dependence of ρ_{AH} and ρ_{xx} for $[\text{Co}/\text{Pd}_{1-x}\text{Ag}_x]_6$ multilayers is shown in Figs. 2(a) and 2(b), respectively. For $x=0$, the negative ρ_{AH} and the decrease in the amplitude of $|\rho_{AH}|$ with increasing temperature have been attributed to surface scattering-dominated AHE.¹³ These characteristics are also presented in the multilayers with $x=0.04$ and 0.19. However, for $x=0.32$ and 0.52, ρ_{AH} is positive, as can be seen clearly from the enlarged view in the inset in Fig. 2(a). ρ_{AH} increases with raising temperature up to 250 K for $x=0.32$ and in the whole temperature range for $x=0.52$. The increase of ρ_{AH} with increasing temperature is a typical behavior of bulk/interface scattering-dominated AHE. The decrease in ρ_{AH} for temperature above 250 K for $x=0.32$ implies the positive AHE from surface scattering, which is suppressed with increasing temperature due to the decrease in mean free path.

The temperature dependence of longitudinal resistivity ρ_{xx} for $[\text{Co}/\text{Pd}_{1-x}\text{Ag}_x]_6$ multilayers is shown in Fig. 2(b), which indicates that all the films are metallic. The overall tendency is that ρ_{xx} decreases with increasing x , except an abnormal increase in the film with $x=0.19$. For this film, it

FIG. 1. The representative Hall loops of $[\text{Co}(3 \text{ \AA})/\text{Pd}_{1-x}\text{Ag}_x(5 \text{ \AA})]_6$ multilayers with $x=0, 0.04, 0.19$, and 0.32 measured at 5 K.

FIG. 2. The temperature dependence of (a) ρ_{AH} and (b) ρ_{xx} for the $[\text{Co}(3 \text{ \AA})/\text{Pd}_{1-x}\text{Ag}_x(5 \text{ \AA})]_6$ multilayers with $x=0, 0.04, 0.19, 0.32$, and 0.52 , respectively. Inset in (a): enlarged view for $x=0.32$ and 0.52 .

also shows a smaller H_c than that of $x=0.04$, in contrast to the increase in H_c with increasing x from 0 to 0.04 (Fig. 1). Therefore, the increase in ρ_{xx} in $x=0.19$ could be related to the interface misfit dislocations.

Shown in Fig. 3 are the representative MR curves of $[\text{Co}/\text{Pd}_{1-x}\text{Ag}_x]_6$ multilayers measured at 5 K with magnetic field applied in the film plane, where $\rho_{\parallel}$ corresponds to the magnetic field parallel to the current direction and $\rho_{\perp}$ for the field perpendicular to the current direction. The multilayer with $x=0$ exhibits an AMR behavior [Fig. 3(a)], and the multilayer with $x=0.04$ presents both AMR and GMR behaviors, where the AMR behavior is embodied in the obvious difference between $\rho_{\parallel}$ and $\rho_{\perp}$ at 5 T [Fig. 3(b)]. For $x=0.52$, the multilayer presents a GMR-dominated behavior [Fig. 3(c)].

GMR ratio and ρ_{AH} as a function of x at 5 K for $[\text{Co}/\text{Pd}_{1-x}\text{Ag}_x]_6$ multilayers with $x=0.04, 0.19, 0.32$, and 0.52 are shown in Figs. 4(a) and 4(b), respectively, where the GMR ratio is defined by $\text{GMR ratio} = [\rho_{\max} - \rho(5 \text{ T})] / \rho(5 \text{ T})$, $\rho_{\max}$ is the maximum ρ_{xx} in $\rho_{\parallel}$, and $\rho(5 \text{ T})$ is the value of $\rho_{\parallel}$ at 5 T. In Fig. 4(a), one can find a sharp increase in GMR ratio as x increases from 0.19 to 0.32, where the sign of AHE crossovers from negative to positive, i.e., from surface scattering-dominated AHE to interface scattering-dominated AHE. It is well known that GMR effect is related to spin-dependent interface scattering in FM/NM multilayers. In GMR theory, the mechanism of GMR and amplitude of GMR ratio are correlated with the mean free path and spin-dependent coefficients at the interfaces for specular reflection, transmission, and diffuse scattering.^{21,22} The gradual substitution of Pd by Ag in our experiment, which modifies these parameters, enables us to observe the transition in magnetoresistance from AMR-dominated to GMR-dominated, correlated with the

FIG. 3. The representative MR curves of $[\text{Co}(3 \text{ \AA})/\text{Pd}_{1-x}\text{Ag}_x(5 \text{ \AA})]_6$ multilayers for (a) $x=0$, (b) $x=0.04$, and (c) $x=0.52$ measured at 5 K with magnetic field applied in the film plane.

FIG. 4. (a) GMR ratio and (b) ρ_{AH} as a function of x at 5 K for $[\text{Co}(3 \text{ \AA})/\text{Pd}_{1-x}\text{Ag}_x(5 \text{ \AA})]_6$, respectively.

transition from surface scattering-dominated to interface scattering-dominated AHE. The theoretical models, which have successfully explained the GMR effect, mainly consider spin-dependent interface scattering in the direction normal to the film plane.^{21,22} How the spin-dependent scattering at the interface in the transverse direction, which induces AHE, is still missing.

The present study indicates, with the increase of the substitution of Pd with Ag at $x \geq 0.32$, the decrease in ρ_{xx} results in an increase in mean free path, which is expected to enhance the contribution of spin-dependent surface scattering to the spin transport, but the contribution of spin-dependent interface scattering is enhanced as well, the latter induces co-occurrence of the enhancement in GMR [Fig. 4(a)] and interface scattering-dominated AHE overwhelming AHE from surface scattering [inset in Fig. 2(a)].

Finally, we carried out scaling analysis of the interface scattering contribution on AHE in the multilayers. In order to eliminate surface scattering contribution on AHE, we fabricated the multilayers with repetition up to 80. The temperature dependence of ρ_{AH} for $[\text{Co}/\text{Pd}_{1-x}\text{Ag}_x]_{80}$ is shown in the inset in Fig. 5. Compared with those of multilayers with $n=6$, as shown in Fig. 2(a), ρ_{AH} in the multilayers with $n=80$ is obviously larger than those with $n=6$, in particular, ρ_{AH} changes from negative to positive for $x=0$, and for $x=0.04$ and 0.19 in temperatures above 150 and 100 K, respectively, which should be due to the reduction in surface scattering contribution to AHE.

It has been observed that the surface scattering contribution to AHE could be ignored in $x=0$, i.e., in $[\text{Co}/\text{Pd}]_{80}$ multilayer.¹³ However, with increasing temperature, the sign change in ρ_{AH} from negative to positive in $x=0.04$ and 0.19 , and downward curvature in $x=0.32$ (inset in Fig. 5), indicate that the surface scattering contribution to AHE is still present in these samples. Scaling analysis would fail without subtraction of the surface scattering contribution from the total AHE.¹³ However, for $x=0.52$, ρ_{AH} is positive, and it increases with increasing temperature (inset in Fig. 5). On the other hand, the relatively large MR ratio of this film

FIG. 5. The plot of $\log R_s$ vs $\log \rho_{xx}$ for the $[\text{Co}(3 \text{ \AA})/\text{Pd}_{1-x}\text{Ag}_x(5 \text{ \AA})]_{80}$ multilayers with $x=0$ and 0.52 . The lines are linear fittings to the data. Inset: The temperature dependence of ρ_{AH} for the $[\text{Co}(3 \text{ \AA})/\text{Pd}_{1-x}\text{Ag}_x(5 \text{ \AA})]_{80}$ multilayers with $x=0, 0.04, 0.19, 0.32$, and 0.52 .

indicates that a strong spin-dependent interface scattering is presented. Therefore, we tried to process the scaling analysis for this sample by Eq. (2). The result of $\log R_s$ vs $\log \rho_{xx}$ is shown in the main panel in Fig. 5. For comparison, that of $x=0$ is also presented. A linear fit for the data of $x=0.52$ leads to $\gamma=3.44 \pm 0.13$, and the film with $x=0$ gives $\gamma=5.71 \pm 0.25$. The difference in the magnetotransport properties in these two multilayers is worth noticing. The sample of $x=0$ exhibits an AMR behavior arising from spin-orbit coupling, while the sample with $x=0.52$ exhibits a GMR behavior. How these distinguished transport properties affect AHE, thus the different γ values, is still not clear.

In summary, we studied AHE and magnetoresistance behavior in $[\text{Co}/\text{Pd}_{1-x}\text{Ag}_x]_n$ multilayers. All these samples present perpendicular magnetic anisotropy. For the multilayers with $n=6$, the AHE sign change from negative to positive has been attributed to the Ag substitution induced enhancement in the interface scattering contribution to AHE, while weakening the surface scattering contribution, though longitudinal resistivity was found to decrease in high Ag content. The enhancement in interface scattering contribution to AHE was associated with the increase of giant magnetoresistance. For the multilayers with $n=80$, scaling analysis yields $\gamma \sim 3.44$ for the sample with $x=0.52$, which exhibits a GMR-type transport behavior. In contrast, $\gamma \sim 5.7$ for the sample with $x=0$ which presents an AMR-type transport behavior.

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