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Temperature and Co thickness dependent sign change of the anomalous Hall effect in Co/Pd multilayers: An experimental and theoretical study

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The anomalous Hall effect in ultra-thin $\{Co^{0.3\,nm}/Pd^{0.5\,nm}\}_n$ multilayers has been investigated recently with respect to surface and interface contributions [Guo *et al.*, Phys. Rev. B **86**, 104433 (2012)]. In this work, we observe a Co thickness and temperature dependent sign change also for $\{Co^{0.20\text{–}0.55\,nm}/Pd^{1.5nm\ \text{or}\ 1.8\,nm}\}_{9x}$ multilayers, e.g., in layer stacks with considerably thicker Pd layers and hence lower resistivity. The thickness dependent behavior can be reproduced by *ab initio* calculations of the Hall conductivity, for which only interfacial and bulk contributions play a role. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4776737]

The anomalous Hall effect (AHE)[1,2] of ultra-thin multilayered (ML) films with large perpendicular magnetic anisotropy (PMA) has recently attracted considerable attention due to its potential applications for high-density magnetic recording technology, such as recording media, magnetic random access memory (MRAM), and magnetic sensors.[3–7]

For applications, parameters like squareness and coercivity of the magnetic hysteresis curve and the amplitude of the AHE voltage are desired to be tunable. For instance, the coercivity has to be as small as possible when it is used for magnetic field sensor elements, but it has to be moderately large when it is used for recording media elements.

The empirical expression of the Hall resistivity is given as $\rho_{xy} = \rho_{OH} + \rho_{AHE} = R_0 \cdot \mu_0 H + R_S M$ (Refs. 8 and 9), where $\rho_{OH} = R_O \cdot \mu_0 H$ represents the ordinary Hall resistivity (R_0 is the ordinary Hall coefficient). The anomalous Hall resistivity $\rho_{AHE} = R_S \cdot M$ is obtained by subtracting a straight line $R_0 \mu_0 H$ from the Hall resistivity ρ_{xy} measured in dependence of H. In this work, the ordinary contribution is small compared to ρ_{AHE} for all samples.

Since Co/Pd is one of the promising candidates for perpendicular magnetic recording technology,[3–5] there are several AHE studies on thin Co-Pd alloys and $\{Co/Pd\}_n$ multilayers. In chronological order, Kim *et al.*[10] explored the Pd thickness dependence of the AHE of {Co/Pd} multilayers prepared by thermal evaporation at room temperature and observed a polarity change of the AHE between 4 and 5 monolayers of Pd for a fixed Co thickness of one monolayer. It was speculated that this can be attributed to a change of the position of the Fermi level with increasing Pd thickness. Jen *et al.*,[11] studied the AHE coefficient (R_S) of $Co_{1-x}Pd_x$ alloys. With increasing Pd concentration x, they observed a sign change from positive to negative at $x = 0.77$ and claimed that the AHE is dominated by the side jump mechanism for $x \leq 0.65$, whereas for $x > 0.65$, both the side jump and skew scattering mechanism are equally effective.

Aoki *et al.*[12] studied {Co/Pd} multilayers deposited from e-gun sources at room temperature. For a constant Pd thickness of 2.0 nm, they found that at 4.2 K, the sign of R_S changes for a Co layer thickness of about $d_{Co} = 0.2$ nm compared to $d_{Co} = 1.6$ nm. Stimulated by the work of Jen *et al.*, they attributed their observation to an interfacial alloying effect. However, they mentioned that they could not rule out the possibility that the sign change was a direct result of the different easy axes orientation for $d_{Co} = 0.2$ nm (showing perpendicular magnetic anisotropy) and $d_{Co} = 1.6$ nm (in-plane anisotropy).

Recent studies on {Co/Pd} multilayers observed a reversal of the AHE polarity both with repetition number[1,13] and aging effects at room temperature[13] in the limit of small Co thickness (0.2 nm in Ref. 13 and 0.3 nm in Ref. 1). This was attributed to a decreasing importance of the surface scattering with increasing number of multilayers compared to the bulk and interface scattering, respectively.

In this work, we report a Co thickness and temperature dependent sign change for $\{Co^{0.20\text{–}0.55\,nm}/Pd^{1.5nm\ \text{or}\ 1.8\,nm}\}_{9x}$ multilayers showing a high PMA with nearly perfect rectangular magnetization switching in the out-of-plane direction and compare the experimental results with *ab initio* calculations of the anomalous Hall conductivity (AHC).

The $\{Co/Pd\}_{9x}$ multilayers with Co thickness ranging from 0.20 nm to 0.55 nm by 0.05 nm steps and a fixed Pd thickness of 1.5 nm or 1.8 nm were grown on thermally oxidized Si wafers by using dc magnetron sputtering. A 5 nm thick Ta buffer layer was deposited prior to the multilayers. The stacks were covered by a 2.1 nm thick MgO layer to prevent oxidation. The samples were annealed at 200 °C for 60 min in a magnetic field of 6500 Oe perpendicular to the film plane to enhance the perpendicular magnetic anisotropy of the {Co/Pd} multilayers. To fabricate the Hall bars, the films were patterned by UV lithography and Ar-ion beam etching to an effective length of L = 1.2 mm and a width of w = 1.0 mm. The Hall voltage V_H (measured in y-direction) and the longitudinal voltage in x-direction (V_x) were driven by a bias current in the x-direction ($I_x = 500\,\mu$A), the

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magnetic field H of up to 10kOe was applied perpendicular to the film plane in z-direction. The low-temperature AHE measurements were done in a closed-cycle helium cryostat (Oxford Cryodrive 1.5) with a temperature range of 15–330 K. The amplitude of the longitudinal magneto resistance R_{xx} (H) = V_x (H)/I_x was observed to be less than 0.5% and will not be further discussed here. The longitudinal resistivity is defined as $\rho_{xx} = (V_x/I_x)(w/L)t$, the Hall resistivity accordingly $\rho_{xy} = (V_H/I_x)t$, where t is the total thickness of the metallic material including 5 nm of Ta.

Sputtered Cobalt-Palladium multilayers have an artificial superlattice structure, where Co and Pd grow textured in the (111) direction and where a compositionally sharp interface can be fabricated.[14] We have confirmed the (111) texture in our films by x-ray diffraction, whereas the in-plane orientation of the grains was random. The according lattice spacings of our films in the growth direction fit very well with the data reported in Ref. 14: 0.2236 nm for d_{Co} = 0.20 nm, 0.2226 nm for d_{Co} = 0.30 nm, and 0.2209 nm for d_{Co} = 0.55 nm (Pd thickness was 1.5 nm in all cases). Furthermore, the Pd atoms at the interface to the Co layer can be expected to be magnetically polarized by the Co atoms.[14]

The thickness dependent room temperature (RT) data of the Hall resistivity ρ_H as a function of the out-of-plane magnetic field H are plotted in Fig. 1 for d_{Co} = 0.20, 0.25, 0.30, and 0.55 nm. It is obvious that the polarity of the AHE loops remains negative for the thin films (0.20 nm and 0.25 nm Co) and that it changes to positive for the thicker ones (0.30 nm to 0.55 nm Co). This behavior is similar to the data reported by Aoki *et al.*[12] However, in our case, the sign change is also observed at room temperature and—because of the high out-of-plane remanence for all multilayers—it can be excluded that the sign change results from a rotation of the easy magnetic axis from out-of-plane to in-plane. The coercive field H_C increases from about 100 Oe for the thinnest film up to 700 Oe (0.30 nm Co) and dropped to 120 Oe with increasing Co thickness, as displayed in the inset of Fig. 1.

The critical (Curie-) temperature T_C for the magnetic phase transition of the $\{Co^{0.20\,nm}/Pd^{1.8\,nm}\}_{9x}$ multilayer is observed at about 300 K as can be seen in Fig. 2(b), however, the AHE resistivity is negative for all temperatures. In contrast for the $\{Co^{0.35\,nm}/Pd^{1.8\,nm}\}_{9x}$ multilayer ρ_{xy} exhibits a temperature dependent sign change between 140 K and 160 K from negative at lower temperatures to positive at higher temperatures. This temperature dependent polarity change is also observed for the $\{Co^{0.30\,nm}/Pd^{1.5\,nm}\}_{9x}$ multilayer with a transition temperature between 120 K and 140 K (see, Figs. 2(a) and 3(b)). However, the nearly perfect squareness of the hysteresis loops is conserved in all measurements, and the normal reduction of H_C with increasing temperature is also observed for all samples.

Similar trends have been observed both for multilayers with 1.8 nm and 1.5 nm Pd (see, Fig. 3(b)), although the critical temperature of the sample with d_{Co} = 0.20 nm and 1.5 nm Pd is also above 330 K and, therefore, larger than in the case of d_{Pd} = 1.8 nm (see, Fig. 1). For this sample, the Hall resistivity ρ_{xy} exhibits a non-monotonic temperature dependence with a broad local minimum around 140 K (see, Fig. 3(b)). For larger Co thickness, a monotonically increasing ρ_{xy} was observed.

In general, the longitudinal resistivity ρ_{xx} increases monotonically with temperature and with a reduction of the film thickness, which is a well known phenomenon for metal thin films[15] and multilayers[16] (see, Fig. 3(a)): the influence of the interface scattering becomes more important if the

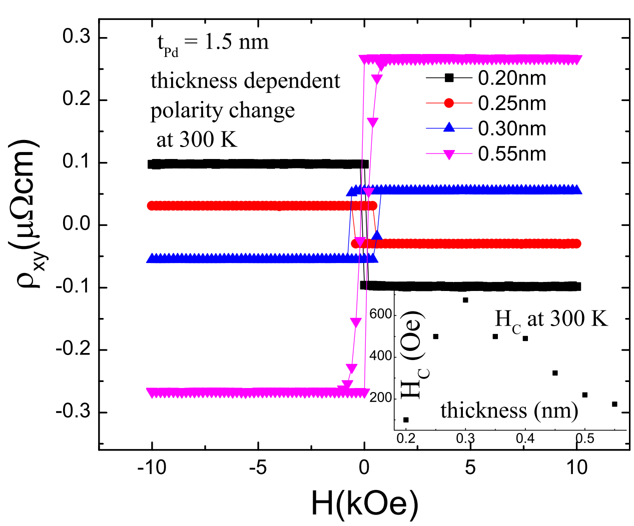

FIG. 1. (a) Hall resistivity as a function of magnetic field for samples of 0.20, 0.25, 0.30, and of 0.55 nm of Co at RT. Inset: The coercivity field H_C of all samples at RT. The Pd thickness was 1.5 nm.

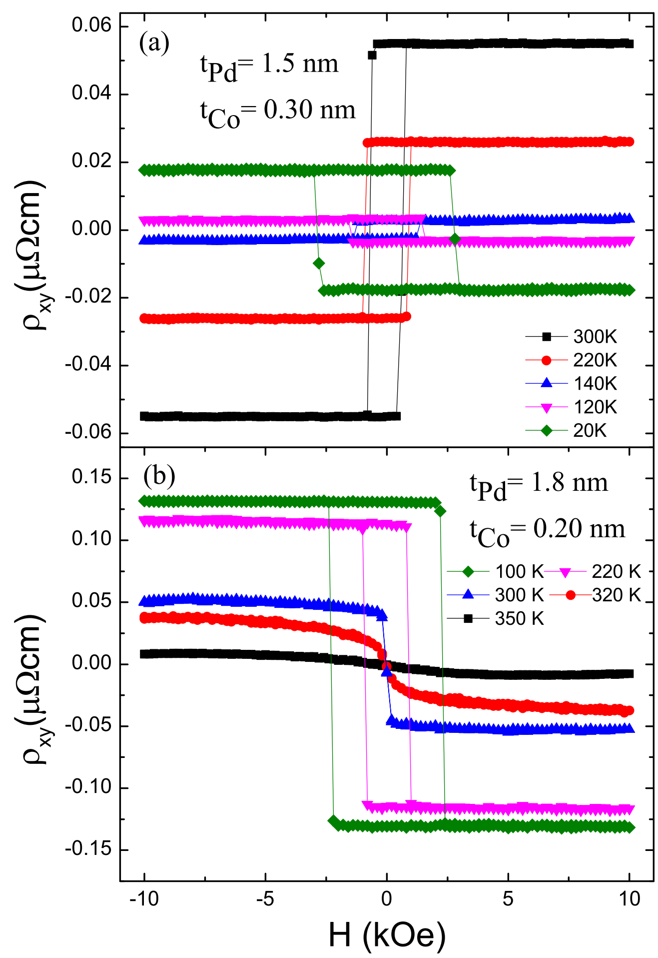

FIG. 2. ρ_{xy} vs. magnetic field H at different temperatures T for (a) d_{Co} = 0.30 nm (the Pd thickness was 1.5 nm) and (b) d_{Co} = 0.20 nm (the Pd thickness was 1.8 nm).

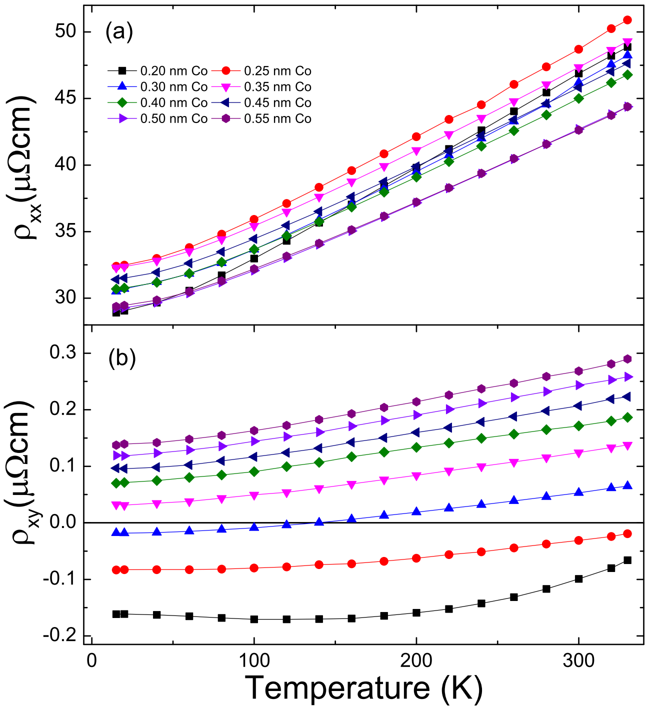

FIG. 3. Temperature dependence of (a) ρ_{xx} and (b) ρ_{xy} for all samples with Pd thickness of 1.5 nm.

number of repetitions and, accordingly, the number of interfaces in the multilayer remains constant while the thickness of one or both single layers is reduced (here the Co thickness is reduced). This is an important hint to a significant influence of the Co/Pd interface on the electrical properties of the multilayers. One, therefore, should also expect a significant effect on the Hall resistivity.

Conventionally, the dependence of the Hall resistivity ρ_{xy} is plotted vs. the longitudinal resistivity ρ_{xx} to distinguish between intrinsic, side-jump and skew scattering contributions.[17] However, in the case of multilayers, this is not always helpful for understanding the physical origin of the temperature dependence of ρ_{xy} because of bulk, interface and surface contributions to both, ρ_{xx} and ρ_{xy}, and an accordingly inhomogeneous current distribution in the stack.[16] Furthermore, all contribution might have different temperature dependencies. But one can only measure two voltages (V_x and V_H) and, therefore, a non-ambiguous separation between all contributions can be hardly done.

Therefore, we have chosen a different approach to understand the experimental low temperature data, where thermal excitations are suppressed. We performed first principles calculations of the electronic properties of {Co/Pd} multilayers with various number of Co (Pd) atomic layers using the full-potential linearized augmented plane wave (FLAPW) code FLEUR[18] and the generalized gradient approximation for the exchange correlation potential.[19] A total number of 288 kpoints in the Brillouin zone (BZ) was used to do self-consistent calculations with spin-orbit coupling. The supercells to simulate the multilayers were constructed assuming fcc stacking of all atomic layers along (111) direction with constant in-plane lattice constants derived from the experimental lattice constant of Pd (3.89 Å). The distance between Co atomic layers (1.7 Å) was taken to be the distance between Co layers as in bulk hcp Co (reduced because of the constant volume approximation), and the distance between Co and Pd layers was taken to be the average of those of Co and Pd layers. The intrinsic AHC is calculated using the Wannier interpolation technique,[20–22] given by the Kubo formula

$$\sigma_{ij} = -e^2\hbar\int_{BZ}\frac{d^3k}{8\pi^3}\Omega_{ij}(\mathbf{k}), \tag{1}$$

$$\Omega_{ij}(\mathbf{k}) = -2\,\mathrm{Im}\sum_{\mathrm{n,m}}^{\mathrm{o,e}}\frac{\langle\psi_{\mathbf{nk}}|\mathrm{v_i}|\psi_{\mathbf{mk}}\rangle\langle\psi_{\mathbf{mk}}|\mathrm{v_j}|\psi_{\mathbf{nk}}\rangle}{(\varepsilon_{\mathbf{nk}}-\varepsilon_{\mathbf{mk}})^2}, \tag{2}$$

which relates the conductivity tensor σ_{ij} (Eq. (1)) to the BZ integral of the **k**-dependent Berry curvature tensor Ω_{ij} (Eq. (2)). In the latter expression, $\psi_{\mathbf{nk}}$ and $\psi_{\mathbf{mk}}$ are, respectively, the occupied (o) and empty (e) eigenstates, $\varepsilon_{\mathbf{nk}}$ and $\varepsilon_{\mathbf{mk}}$ are their eigenenergies, and $\mathrm{v_i}$ and $\mathrm{v_j}$ are the Cartesian components of the velocity operator **v**.

Fig. 4 displays the AHC of {Co/Pd} multilayers with various composition calculated using the first principles methods. We considered here the intrinsic contribution given by the Kubo formula above. Obviously, the AHC varies significantly with respect to the location of the Fermi energy E_F due to the fact that the Berry curvature Ω is a sensitive quantity. For instance, the AHC of Co_3Pd_6 multilayers oscillates between positive and negative values. This can cause a nontrivial temperature dependence of the intrinsic AHC. However, to compare with experimental observations at low temperature, the AHC around E_F is relevant. As shown in the inset of Fig. 4, the AHC at E_F has a strong dependence to the number of Co atomic layers. The sign of AHC is changed from negative to positive for multilayers with more than 2 Co layers (0.34 nm). This is in agreement with our experimental results that at low temperature, the sign of ρ_{xy} is changed at a critical thickness of 0.30 ∼ 0.35 nm (see, Fig. 3(b)).

In order to roughly estimate the magnitude of the extrinsic contribution to the AHE in the system, we additionally

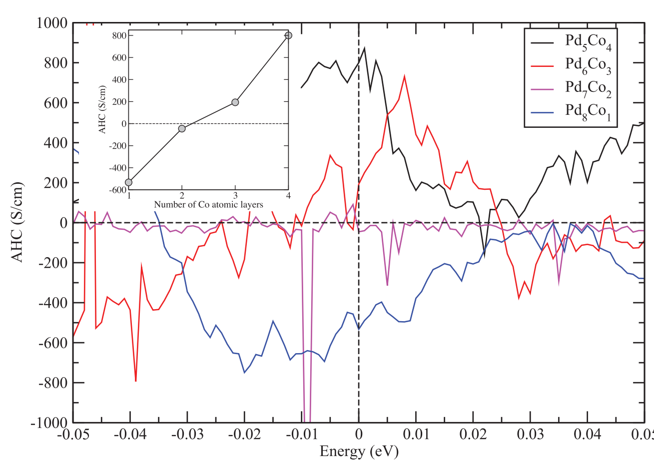

FIG. 4. Anomalous Hall conductivities of Co_mPd_n multilayers. Compositions with m = 1, 2, 3, 4 and corresponding n = 8, 7, 6, 5 are considered with 9 atomic layers in total to maintain fcc stacking. Horizontal (vertical) dashed line indicates the zero AHC (location of the Fermi energy). The inset displays the dependence of the AHC at the Fermi energy E_F with respect to the number of Co atomic layers. The solid line is the guide for the eyes.

performed an *ab intio* assessment of the side-jump contribution, which can have a comparatively large magnitude in transition-metals, see, e.g., Refs. 23–25. Since the exact details of disorder in the system are unknown and the skew-scattering contribution to the AHE becomes increasingly suppressed as the temperature of the sample is increased in transition-metals,[17] we choose a model of Gaussian disorder to estimate the so-called scattering-independent side-jump contribution, known to be important in such metals as Fe, Co, and FePd.[25] Our zero-temperature calculations performed according to the methodology of Ref. 25 show that the magnitude of the side-jump in Co/Pd multilayers does not exceed the magnitude of the intrinsic contribution and has the same sign as the intrinsic values. Noticably, while for the Co_1Pd_8 multilayer, the values of both side-jump and intrinsic contributions are negative, they converge to the bulk Co values[25] as the number of the Co layers in the system is increasing. The overall trend with increasing Co thickness, which is in agreement with experimental observations, provides a hint that the sign change in the anomalous Hall signal is caused by a competition between bulk Co and interfacial contribution, with the spin-polarization of the interfacial Pd atoms possibly playing an important role for the latter.

Now, we come back to the temperature dependence of ρ_{xy}. Although temperature dependent calculations from first principles are challenging in general, the low temperature calculations could be qualitatively analyzed with respect to a potential temperature dependence in a crude approximation as follows: assuming a constant magnetization, the temperature dependence of the AHC is estimated by averaging it in the neighborhood of the Fermi energy E_F. Since the energy behavior of the side-jump contribution is rather smooth as compared to the intrinsic, it can be ignored for the estimate of the temperature dependence of the AHE. Thus, considering the intrinsic AHC in the $+/-0.03$ eV region around E_F, which roughly corresponds to 300 K, the averaged AHC for Co_1Pd_8 would be always negative, while that for Co_4Pd_5 would be always positive for $T \leq 300$ K. Nevertheless, for Co_3Pd_6 multilayers, the averaged AHC would decrease and could change its sign due to negative contributions for $|E| \geq 0.025$ eV. This effect is not observed experimentally, however, as shown in Fig. 3(b), and the general trend is an increasing ρ_{xy} with temperature. This means that for the temperature dependent considerations, it is not sufficient to take only the intrinsic and scattering-independent side-jump AHC into account by averaging it with respect to the energy and to ignore contributions to the AHE, which depend on the details of disorder at the interface and in the bulk. Although the individual temperature dependencies of the different contributions cannot be extracted from our data because of averaging over the whole sample, the limit of largest Co thickness shows that with increasing temperature the bulk at least gives rise to an increasing positive contribution in ρ_{xy} in our {Co/Pd} multilayers with thin Co layers and relatively large Pd thickness. Assuming that scattering at the interface is effectively a scattering at an impurity (in first order the temperature dependence of this scattering can be ignored) and taking into account that the bulk resistivity of Co (and Pd as well) increases with increasing temperature because of electron-phonon scattering or other inelastic excitations (e.g., magnons), it is reasonable to assume that the positive bulk contribution to ρ_{xy} increases with temperature because of scattering dependent contributions to the AHE.

To summarize, we report on the Co thickness and temperature dependence of the Hall resistivity in $\{Co^{0.20-0.55\text{nm}}/Pd^{1.5\text{nm or }1.8\text{nm}}\}_{9x}$ multilayer systems. In particular, a change of its sign was observed both in the dependence of ρ_{xy} on the Co thickness and on the temperature. We have shown that the low temperature data can be understood on the base of calculations from first principles, which take only scattering-independent contributions into account. Moreover, it has been shown that the simplest possible evaluation of the temperature dependence from these calculations is not enough to explain the experiments. However, taking the temperature dependence of ρ_{xy} of the multilayers with largest Co thickness into account, it can be argued that the general increase of ρ_{xy} with temperature is significantly influenced by the temperature dependence of the (positive) bulk contribution via phonons, magnons, etc. Furthermore, in the case of large Pd thickness investigated here, surface scattering seems to be not as important as in multilayer systems with significantly smaller Pd thickness as reported in Ref. 1.

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