

**Absence of spin Hall magnetoresistance in Pt/(CoNi)<sub>n</sub> multilayers**Yongwei Cui<sup>1</sup>,<sup>✉</sup> Xiaoyu Feng,<sup>1</sup> Qihan Zhang,<sup>1</sup> Hengan Zhou,<sup>2</sup> Wanjun Jiang,<sup>2</sup> Jiangwei Cao<sup>1</sup>,<sup>✉</sup> Desheng Xue,<sup>1</sup> and Xiaolong Fan<sup>1,\*</sup><sup>1</sup>Key Laboratory for Magnetism and Magnetic Materials of the Ministry of Education, Lanzhou University, Lanzhou 730000, People's Republic of China<sup>2</sup>State Key Laboratory of Low-Dimensional Quantum Physics and Department of Physics, Tsinghua University, Beijing 100084, China (Received 26 July 2020; revised 21 October 2020; accepted 10 December 2020; published 11 January 2021)

We have systematically studied the magnetoresistance effect in Pt/(CoNi)<sub>n</sub> multilayers featured with perpendicular magnetic anisotropy and fcc (111) texture. Angular dependence of the magnetoresistance, including high-order cosine terms, was found in the plane perpendicular to the electrical current, which can be attributed to the geometrical size effects and the anisotropic interface magnetoresistance effect. However, the contribution from spin Hall magnetoresistance (SMR) is absent. The spin Hall angle of Pt was evaluated by spin-torque ferromagnetic resonance measurements, and the theoretical magnitude of SMR in our samples was estimated to be one order higher than the accuracy of our experiments. The absence of SMR in our multilayers indicates that the Elliott-Yafet spin relaxation of itinerant electrons in the ferromagnetic metal is crucial when studying the spin transport in all-metallic heterostructures.

DOI: [10.1103/PhysRevB.103.024415](https://doi.org/10.1103/PhysRevB.103.024415)**I. INTRODUCTION**

Spin Hall magnetoresistance (SMR) refers to the anisotropic absorption of spin current with respect to the relative orientation of magnetization in the heavy-metal (HM)/ferromagnet bilayers, by a change in HM resistance with the direction of magnetization [1–3]. Because SMR essentially reflects the interaction between spin current and local magnetic moment, it has become a powerful tool for studying spin-orbit coupling in magnetic heterostructures. For examples, SMR can be used to determine the spin Hall angle, spin-diffusion length in HM, and interface spin-mixing conductance [2–5], even interfacial spin-orbit coupling can be studied by spin-orbit MR [6]. In addition, SMR can also be used to monitor the directions of the Néel order in antiferromagnetic materials by means of highly sensitive electrical measurements [7–10].

An SMR model was first proposed in the heterostructure of HMs and ferromagnetic insulators (FMI) [2,11,12], and was then adopted to study the spin-dependent transport behaviors in all-metallic heterostructures [4,5,13]. Compared with HM/FMI, the SMR effect in an HM/ferromagnetic metal (FM) bilayer is more complicated. Based on previous theoretical proposals, the absorption of the longitudinal spin current by the FM layer [5], the spin current generated by the FM layer [14], and the anomalous Hall effect of the FM layer [4,15,16] should also be considered. More importantly, although the appearance of the magnetoresistance effect in the plane perpendicular to the charge current (i.e., the *yz* plane) can be used to distinguish SMR from traditional anisotropic magnetoresistance (AMR), the ordered arrangement of grains in

FMs and the interfacial symmetry breaking could also lead to the appearance of the magnetoresistance effect in that plane [17–19]. For example, Kobs *et al.* observed distinct magnetoresistance in the *yz* plane in a Pt/Ni/Pt system, but found that the contribution of SMR could almost be ignored [20]. The absence of SMR in all-metallic heterostructure brings the other complicity which implies either the spin-orbit coupling is quite weak, or the SMR model in HM/FMI cannot be directly adopted. Hence, it is necessary to revisit the magnitude of SMR in HM/FM bilayers.

In the present paper, we employed (CoNi)<sub>n</sub> multilayers with perpendicular magnetic anisotropy (PMA) to study SMR effect. The difference from other films with PMA such as Ta/CoFeB/MgO is their artificially tunable PMA over a significantly wider range of magnetic thickness [21–23]. Therefore, (CoNi)<sub>n</sub> multilayers are desirable spintronics materials to study magnetic domain-wall motion and magnetization switching [9,24–26]. In addition, (CoNi) multilayers exhibit moderate intrinsic damping and higher spin polarization [27–29] compared with other bulk PMA films, enabling to develop better performance spin-torque magnetic random access memories (MRAMs) and data storage applications [30]. However, we found that the magnitude of SMR in Pt/(CoNi)<sub>n</sub> is far less than what was predicted theoretically based on the measurements of spin-torque ferromagnetic resonance (ST-FMR), which led us to further explore the mechanism of spin transport in all-metallic magnetic heterostructures.

**II. THIN-FILM DEPOSITION AND STRUCTURAL CHARACTERIZATION**

In this work, Ta(3)/Pt(10)/[Co(0.3)/Ni(0.4)]<sub>n</sub>/Ta(3) were deposited onto SiO<sub>2</sub>/Si substrate by magnetron sputtering at room temperature. The numbers in brackets represent

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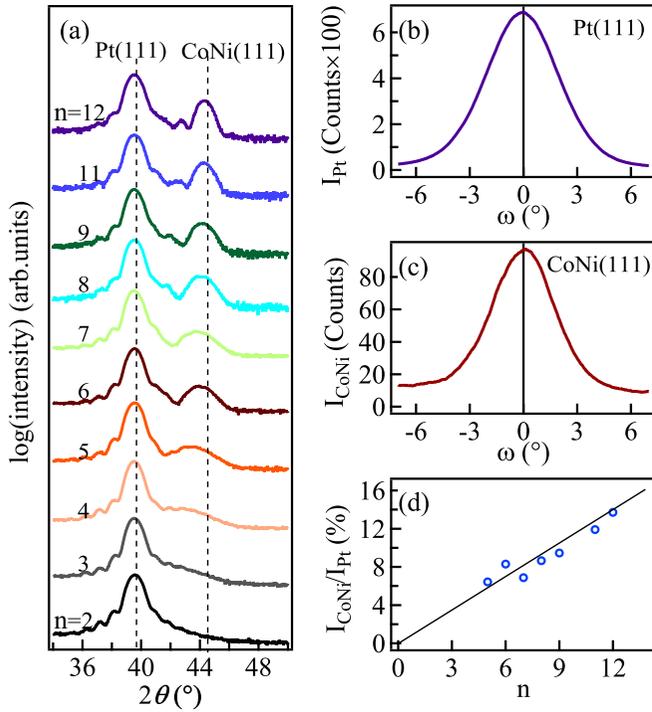


FIG. 1. (a) Out-of-plane x-ray measurement of Ta(3)/Pt(10)/[Co(0.3)/Ni(0.4)]<sub>n</sub>/Ta(3) for  $n$  ranging from 2 to 12. The dashed lines at  $39.7^\circ$  and  $44.5^\circ$  represent the expected positions of unstrained Pt (111) and CoNi (111) peaks, respectively. (b), (c) Rocking curves around Pt (111) and [Co/Ni]<sub>12</sub> (111). (d) The blue cycles are the dependence of the peak intensity ratio of [Co/Ni]<sub>n</sub> and Pt on the number of repetitions  $n$ . The black line is a straight line through the origin that was fitted to our results.

the thickness of each layer in nanometer units.  $n = 2, 3, 4, 5, 6, 7, 8, 9, 11,$  and  $12$  are the number of repetitions of the magnetic double layer. The thickness of Pt, Co, and Ni layers is the optimized choice that the samples show maximum PMA at a certain repetition number  $n$ . Moreover, Pt/Co/Ni stacking order is also favorable to obtain perpendicularly magnetized CoNi multilayers due to a larger interface anisotropy at Pt/Co interface compared with Pt/Ni [31]. The Ta in the lower and upper layers form buffer and protective layers, which reduce the influence of substrate roughness on the film and prevent the film from being oxidized. The base pressure of the main chamber is better than  $2 \times 10^{-8}$  Torr, and the Ar pressure is maintained at  $3 \times 10^{-3}$  Torr during sputtering. The static magnetic properties of all the samples were obtained by vibrating-sample magnetometry, and the crystal structure was characterized by high-resolution x-ray diffractometry (HR-XRD) with a Cu  $K\alpha$  radiation source. The magnetoresistance was characterized using the physical property measurement system (Quantum Design), and ST-FMR test was completed using a home-built system.

First, we determined the crystal structure and orientation of all the samples. Figure 1(a) shows the  $\theta - 2\theta$  x-ray-diffraction spectrum of samples with different repetition numbers, where the dashed lines at  $39.7^\circ$  and  $44.5^\circ$  represent the unstrained positions of the Pt (111) and CoNi (111) peaks, respectively [32,33]. For  $n = 2$ , only Pt (111) peak appears in the range of

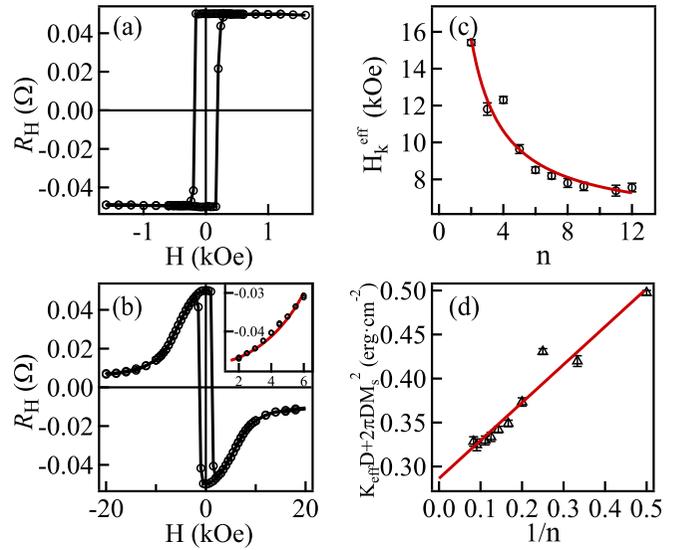


FIG. 2. (a), (b) The loops of abnormal Hall resistance when a magnetic field was applied along the film normal and plane of  $n = 8$ . The inset in (b) shows local enlarged data from 1.6 to 6 kOe, and the curve is the result of fitting using Eq. (1). (c) The black circles represent the dependence of the effective PMA field on the repetition number, which can be well fitted by  $A/n + B$  ( $A$  and  $B$  are constants) as the curve shows. (d) The triangle represents the perpendicular anisotropy energy per unit area, and the red line denotes a linear fitting.

$34^\circ$ – $50^\circ$ , and a number of clear secondary diffraction peaks generated by Laue oscillation can be seen around this peak, indicating that Pt has a high crystallinity quality in the (111) orientation and low interface roughness. When  $n > 4$ , a relatively weak peak appears at  $42^\circ$ – $44^\circ$ . As  $n$  increases, the strength of the peak gradually becomes stronger and the peak position gradually approaches  $44.5^\circ$ . Using Gaussian fitting, the full width at half maximum of the rocking curves around the Pt (111) and CoNi (111) reflections can be determined to be  $5.7^\circ$  and  $4.9^\circ$ , respectively, as shown in Figs. 1(b) and 1(c). This indicates that the Pt and CoNi in the samples have (111) texture along the normal direction of the film. In order to illustrate the precise control of the number of repetition, the ratio of the intensity of CoNi (111) and Pt (111) is shown in Fig. 1(d). It is evident that a straight line through the zero point can be used to fit our experimental results (open circles). Thus, changing the number of repetitions only increases the number of crystallographic plane participating in Bragg reflection, but does not affect the crystal structure of each CoNi layer.

To further demonstrate that we have achieved accurate regulation of CoNi repetition, we also measured the dependence of PMA on  $n$ . As shown in Figs. 2(a) and 2(b), the abnormal Hall resistance curve of  $n = 8$  was measured when a magnetic field was applied out of plane and in plane. The presence of PMA is evident from square-shaped anomalous Hall effect (AHE), as shown in Fig. 2(a) [22,34]. In order to obtain the exact value of  $H_k^{\text{eff}}$ , we use the following formula [35]:

$$\frac{\rho_{xy}(H_x)}{\rho_{xy}(0)} = \sqrt{1 - \left(\frac{H_x}{H_k^{\text{eff}}}\right)^2} \quad (1)$$

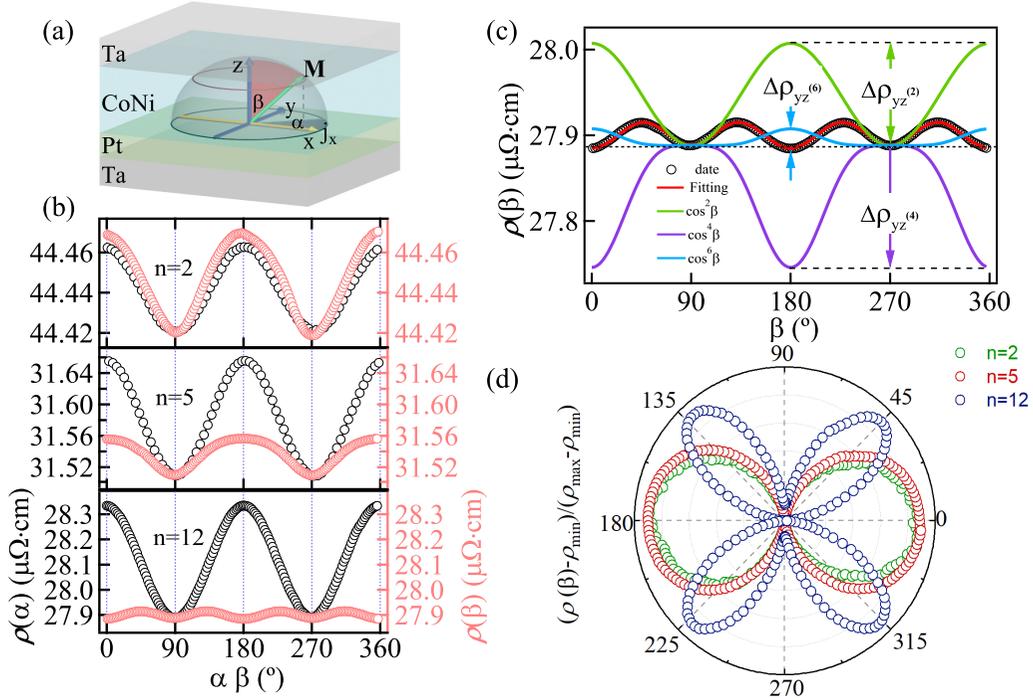


FIG. 3. Magnetoresistance curves of  $\text{Pt}/[\text{Co}/\text{Ni}]_n$  structures. (a) Illustration of the  $\text{Ta}/\text{Pt}/[\text{Co}/\text{Ni}]_n/\text{Ta}$  sample structure and coordinate system of the experiment. (b) Resistivity  $\rho$  vs orientation of magnetization for three samples with different repetitions. The rotation of magnetization  $\mathbf{M}$  is performed in the film plane ( $\beta = 90^\circ$ , black cycles) and in the plane perpendicular to the current  $\mathbf{j}$  ( $\alpha = 90^\circ$ , red cycles). (c) Individual  $\cos^{2n}\beta$  contributions to  $\rho(\beta)$  for  $n = 12$ . (d) Polar plot of  $\rho(\beta)$  for these three samples.

to fit the AHE curve in Fig. 2(b) with the range of 1.6 ~ 6 kOe. The fitting result is  $H_k^{\text{eff}} = 7.6 \pm 0.1$  kOe. Subsequently, the relationship between  $H_k^{\text{eff}}$  and  $n$  can be obtained, as shown in Fig. 2(c). The evolution of  $H_k^{\text{eff}}$  as a function of layer repetition  $n$  suggests dominated interfacial contribution to the strength of PMA [32]. Combined with the method of You *et al.* [22], neglecting the Co and Ni volume anisotropy energies, the average perpendicular anisotropic energy per unit area of each CoNi layer can be written as

$$K_{\text{eff}}D + 2\pi DM_s^2 = K_s^{\text{Co/Ni}} + K_s^{\text{Ni/Co}} + (1/n)(K_s^{\text{Pt/Co}} + K_s^{\text{Ni/Ta}} - K_s^{\text{Ni/Co}}). \quad (2)$$

In this equation,  $K_{\text{eff}} = \frac{H_k^{\text{eff}} M_s}{2}$  is the effective PMA energy,  $M_s = (M_s^{\text{Co}} t_{\text{Co}} + M_s^{\text{Ni}} t_{\text{Ni}})/D = 614.3 \text{ emu cm}^{-3}$  is the saturation magnetization,  $D = 0.7 \text{ nm}$  is the bilayer thickness,  $K_s^{\text{Co/Ni}}$ ,  $K_s^{\text{Ni/Co}}$ ,  $K_s^{\text{Pt/Co}}$ , and  $K_s^{\text{Ni/Ta}}$  are the interface anisotropy energies of the Co/Ni, Ni/Co, Pt/Co, and Ni/Ta interfaces, respectively. As shown in Fig. 2(d), there is a linear relationship between this energy and  $1/n$ . Given that  $K_s^{\text{Co/Ni}} = K_s^{\text{Ni/Co}}$  and neglecting the  $K_s^{\text{Ni/Ta}}$  term,  $K_s^{\text{Co/Ni}} = 0.14 \pm 0.01 \text{ (erg cm}^{-2}\text{)}$  and  $K_s^{\text{Pt/Co}} = 0.57 \pm 0.04 \text{ (erg cm}^{-2}\text{)}$  can be obtained. These values are close to what has been previously reported [22,36]. Thus, based on the above-mentioned analysis of crystal structure and interface anisotropy, the samples exhibit clear periodic structures and Co/Pt and Co/Ni interfaces.

### III. ANGLE DEPENDENCE OF THE MAGNETORESISTANCE

It is generally believed that the presence of second-order cosine symmetric magnetoresistance in the HM/FM structure in the plane perpendicular to the direction of current [i.e., the  $yz$  plane as defined in Fig. 3(a)] is a sign of the SMR [1]. However, in order to strictly separate the contributions of SMR from the geometrical-size effect (GSE) [37,38] and anisotropic interface magnetoresistance (AIMR) [17,18], it is necessary to obtain the thickness dependence of the magnetoresistance in both the  $xy$  and  $yz$  planes. (See Supplemental Material [39], S3.)

As shown in Fig. 3, we measured the angle-dependent magnetoresistance in the  $xy$  and  $yz$  planes. A magnetic field of 9 T is applied to ensure that the magnetic moment of the FM is parallel to the magnetic field. Here,  $\alpha$  and  $\beta$  denote the angle of the magnetic field direction measured from the  $x$  axis in the  $xy$  plane and from the  $z$  axis in the  $yz$  plane. The experimental results of three typical samples are shown in Fig. 3(b). In particular, a  $\cos^2\alpha$  behavior is found for all CoNi thicknesses in the  $xy$  plane [black circle, Fig. 3(b)], which is consistent with the behavior of conventional AMR. However, the magnetoresistance curve in the  $yz$  plane contains higher-order cosine terms and the contribution of this term increases with  $n$ . The polar diagram in Fig. 3(d) can be used to further highlight the symmetry of the magnetoresistance in the  $yz$  plane. Then, we fit the magnetoresistance curve of the  $xy$  plane using the classical AMR equation [33]:

$$\rho(\alpha) = \rho_0 + \Delta\rho_{xy} \cos^2 \alpha, \quad (3)$$

where  $\rho_0$  refers to the resistivity when the magnetic field is perpendicular to the current direction, and  $\Delta\rho_{xy}$  refers to the change in the resistivity when the magnetic field is aligned with the  $x$ - and  $y$  axes, respectively. A Fourier analysis reveals that orders of  $\cos^{2n}\beta$  have to be considered to properly describe the curves of the  $yz$  plane [20]:

$$\rho(\beta) = \rho_0 + \sum_{n=1}^3 \Delta\rho_{yz}^{(2n)} \cos^{2n}\beta. \quad (4)$$

This fits the experimental data well, as shown in Fig. 3(c). For all the samples, the sixth-order ( $\cos^6\beta$ ) is much smaller than the other two terms, and therefore, it is omitted from the following discussion.

It is certain that there are various magnetoresistance mechanisms in the Pt/[Co/Ni] $_n$  multilayers. To model the system, we assume that the resistance change due to SMR occurs in Pt whereas the change caused by AIMR and GSE takes place within the CoNi layer. Considering the current shunting effect (see Supplemental Material [39] S1), we can attribute all the magnetoresistance effects in the sample to the contribution of a single magnetic layer. Consequently, different magnetoresistance mechanisms can be identified from the relationship between magnetoresistance and the thickness of the magnetic layer [18,20]. Based on this approach, we consider the shunting effect according to

$$\frac{\Delta\rho_{\text{CoNi}}}{\rho_{\text{CoNi}}} = \frac{\Delta\rho}{\rho} \times \frac{d_{\text{Pt}}\rho_{\text{CoNi}} + t_{\text{CoNi}}\rho_{\text{Pt}}}{t_{\text{CoNi}}\rho_{\text{Pt}}}, \quad (5)$$

where  $\frac{\Delta\rho}{\rho}$  is the total magnetoresistance, and  $\rho_{\text{CoNi}} = 1.8 \times 10^{-7} \Omega \text{ m}$  and  $\rho_{\text{Pt}} = 2 \times 10^{-7} \Omega \text{ m}$  represent the resistivity of CoNi and Pt.

The SMR effect is derived from the absorption and reflection of the interface spin current in the Pt layer, which is related to the direction of FM magnetization and is reflected as a magnetoresistance under the combined actions of spin Hall effect (SHE) and inverse SHE. There are three basic characteristics of this effect. First, variation of the magnetoresistance depends on the angle between magnetization and spin polarization ( $y$  axis), so it can be observed in both the  $yz$  and  $xy$  planes. Second, according to the previous theory [1], there is only a  $\cos^2\beta$  term. Finally, as to what we want to highlight, when the SMR is attributed to the magnetoresistance of the CoNi layer, it will show an inverse relationship with the thickness of the CoNi layer. As shown in Fig. 4(a), the magnetoresistance ratio in the  $xy$  plane ( $\frac{\Delta\rho_{xy,\text{CoNi}}}{\rho_{\text{CoNi}}}$ ) shows a linear dependence with the thickness of  $t_{\text{CoNi,corr}}$  and is not inversely proportional to the thickness as we expected. Note that  $t_{\text{CoNi,corr}} = t_{\text{CoNi}} - 0.65 \text{ nm}$  is the effective thickness of the magnetic layer, where the presence of magnetic dead layers (0.65 nm) at the interface between CoNi and Pt has been considered. (See Supplemental Material [39], S2.) Therefore, this result indicates that SMR may be absent in our samples. Nevertheless, we do observe magnetoresistance in the  $yz$  plane, which is usually the basis used to identify SMR. Therefore, we need to further analyze the magnetoresistance observed in the  $yz$  plane.

Figure 4(b) shows a nonmonotonic change between the second-order magnetoresistance ratio in the  $yz$  plane

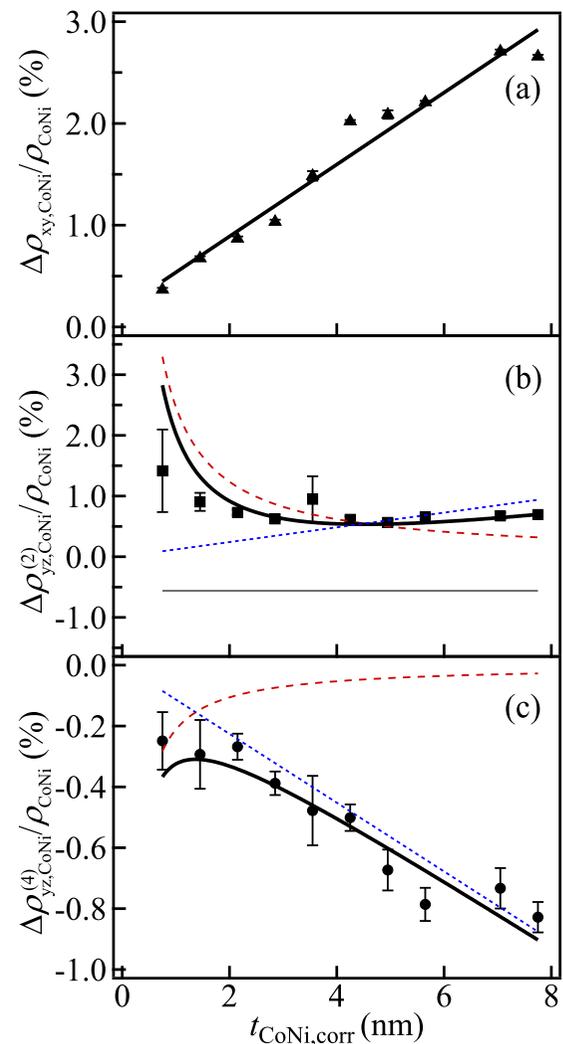


FIG. 4. The thickness-dependent magnetoresistance after corrections of dead layer. (a)  $\Delta\rho_{xy,\text{CoNi}}/\rho_{\text{CoNi}}$  (b)  $\Delta\rho_{yz,\text{CoNi}}^{(2)}/\rho_{\text{CoNi}}$ , and (c)  $\Delta\rho_{yz,\text{CoNi}}^{(4)}/\rho_{\text{CoNi}}$  as a function of the thickness of CoNi. The symbols represent experimental data, while solid black lines represent fitting lines. The dashed red lines represent the contribution of AIMR [the contribution of  $A$  in Eq. (6)]. The GSE contribution is a constant that is independent of the thickness [the contribution of  $C$  in Eq. (6)], which is indicated by a thinner solid straight line in (b). The blue dotted lines indicate the linear contribution, which corresponds to  $B$  in Eq. (6).

( $\Delta\rho_{yz,\text{CoNi}}^{(2)}/\rho_{\text{CoNi}}$ ) and  $t_{\text{CoNi,corr}}$ . When  $t_{\text{CoNi,corr}} < 4 \text{ nm}$ , the magnetoresistance ratio decreases as the thickness increases, thus confirming the existence of an interfacial contribution. When  $t_{\text{CoNi,corr}} \geq 4 \text{ nm}$  the magnetoresistance ratio tends to increase with the thickness. Therefore, we fit the thickness dependence of the magnetoresistance ratio using the following empirical formula:

$$\frac{\Delta\rho_{yz,\text{CoNi}}^{(2n)}}{\rho_{\text{CoNi}}} = \frac{A}{t_{\text{CoNi,corr}}} + Bt_{\text{CoNi,corr}} + C, \quad (6)$$

where three undetermined parameters, namely  $A$ ,  $B$ , and  $C$ , are introduced.  $A$  describes the contribution of the interface,  $B$  represents the contribution of the linear relationship with

the thickness, and  $C$  represents the contribution independent of the thickness. As shown by the black curve in Fig. 4(b), the fitting formula matches the experimental data, and the fitting results are  $A = 0.025$  nm,  $B = 0.012$  nm<sup>-1</sup>, and  $C = -0.0006$ . Assuming that  $A$  entirely comprises SMR contributions, a small percent change in magnetoresistance in the  $xy$  plane should have been observed (e.g.,  $\frac{\Delta\rho_{xy,CoNi}}{\rho_{CoNi}}$  should be 2.5% at  $t_{CoNi,corr} = 1$  nm). However, as shown in Fig. 4(a), the magnetoresistance is below 1% when the thickness is less than 2 nm, which further proves that the contribution of SMR can be neglected.

Three primary components contribute to the magnetoresistance of Pt/(CoNi)<sub>*n*</sub> in the  $yz$  plane. First, AIMR arises because of the anisotropic interfacial scattering of conductive electrons, which only presents when magnetization varies in the  $yz$  plane with high-order cosine terms as shown in the red dashed lines in Figs. 4(b) and 4(c). Second, the negative constant term in the second-order term can be attributed to the GSE effect as indicated by a solid straight line in Fig. 4(b), which is caused by the crystallinity and the anisotropic orientation of the grains [18,20]. Third, the part where there is a linear relationship with the thickness appears in the second-order and fourth-order terms [the blue dotted lines in Figs. 4(b) and 4(c)], and the mechanism for this is still being investigated. In summary, through the above detailed analysis, we proved that SMR was not evident in the samples. Moreover, the presence of a  $\cos^2\beta$  magnetoresistance in the  $yz$  plane is not the only criterion for determining the existence of SMR.

#### IV. THEORETICAL PREDICTION OF THE AMPLITUDE OF SMR

To further investigate the absence of SMR, the theoretical value of SMR in the sample needs to be determined. It is well known that when the Pt layer thickness is much larger than its spin-diffusion length, the spin Hall angle ( $\theta_{SH}$ ) is the only key parameter that determines the SMR value [4]. Therefore, based on spin-torque ferromagnetic resonance methods [14,29,42–45], we introduced a microwave signal with a GHz frequency into the Pt/(CoNi)<sub>*n*</sub> microstrip. The measured typical ST-FMR spectrum is shown in Fig. 5(a), and the experimental conditions are 18.5 GHz, 25 dBm, and  $\alpha = 45^\circ$ . The spectrum can be well fitted with a general line-shape equation as follows:

$$V_{dc} = U_s \frac{\Delta H^2}{\Delta H^2 + (H - H_0)^2} + U_a \frac{\Delta H(H - H_0)}{\Delta H^2 + (H - H_0)^2}, \quad (7)$$

where  $U_s$  and  $U_a$  are the voltage amplitudes of the symmetric Lorentz and antisymmetric dispersive line shapes, which, according to the theory of Sankey *et al.*, are due to the dampinglike torque caused by SHE of the Pt layer and torque caused by the Oersted field [43], and  $\Delta H$  and  $H_0$  are the linewidth and resonance field. In order to accurately obtain the spin Hall angle, we first obtained the angular dependence of  $U_s$  and  $U_a$ , as shown in Fig. 5(b). They all satisfy the theoretical angle-dependence relationship of  $\sin 2\alpha \cos \alpha$ , which means that the ratio between  $U_s$  and  $U_a$  is independent of the angle. (See Supplemental Material [39], S4 for the details

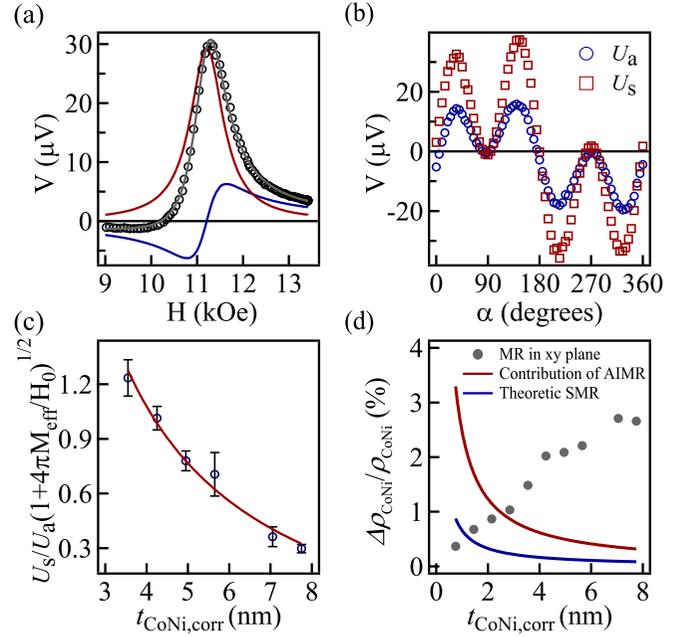


FIG. 5. (a) ST-FMR spectrum of  $n = 7$  sample measured at  $45^\circ$  in plane with a microwave injection of 18 GHz, which includes antisymmetric (blue curve) and Lorentz symmetric (red curve) line shapes. (b) Angular dependence of  $U_s$  and  $U_a$  at 18 GHz in the  $xy$  plane. (c) Fitting the spin Hall angle ( $\theta_{SH}$ ) according to Eq. (9). (d) The dependence of the  $xy$  plane magnetoresistance, the theoretical value of SMR, and the experimental value of AIMR on the thickness of CoNi are put together for comparison.

of the angular-dependent line-shape fitting.) In addition, we measured the  $U_s/U_a$  ratio of samples with different CoNi thicknesses at 18 GHz and then fitted according to Eq. (3) in Ref. [43],

$$\frac{U_s}{U_a} \left[ 1 + \left( \frac{4\pi M_{eff}}{H_0} \right) \right]^{1/2} = \theta_{SH} \frac{\hbar}{\epsilon\mu_0 M_s t_{CoNi,corr} d_{Pt}}. \quad (8)$$

Through the reciprocal relationship with  $t_{CoNi,corr}$ , the spin Hall angle of Pt was directly determined to be 0.07. It should be noted that we considered the interface to be transparent to the spin current generated in Pt. If the transparency of the Pt/Co interface  $T = 0.65 \pm 0.06$  is considered, according to Ref. [46], the spin current density of Pt flowing to the Co/Pt interface is greater than that flowing into the CoNi layer. Thus, the spin Hall angle in our sample is estimated to be larger than 0.07. The SMR  $\sim 7.3 \times 10^{-4}$  estimated by

$$\frac{\Delta\rho_{SMR}}{\rho_{Pt}} \sim \theta_{SH}^2 \frac{\lambda_{Pt}}{d_{Pt}} \tanh\left(\frac{d_{Pt}}{2\lambda_{Pt}}\right) \left[ 1 - \frac{1}{\cosh\left(\frac{d_{Pt}}{\lambda_{Pt}}\right)} \right] \quad (9)$$

should be the theoretical minimum (see Supplemental Material [39], S5) where  $\lambda_{Pt} = 1.5$  nm is the spin-diffusion length of Pt [3].

In order to directly compare the theoretical SMR value with our experimental results, the theoretical SMR value is attributed to the magnetoresistance of the CoNi layer through shunt treatment (see Supplemental Material [39], S1). The theoretical SMR contribution, which is inversely proportional

to the thickness of the CoNi layer in the  $xy$  and  $yz$  planes, corresponds to  $A_{\text{SMR}} \approx 6.57 \times 10^{-3}$  nm in Eq. (6). In order to facilitate comparison, the theoretical prediction of SMR, the second-order contribution of AIMR in the experimental data, and the experimental results of the magnetoresistance in the  $xy$  plane were all included in Fig. 5(d). It is evident that the theoretical SMR is higher than the experimental value, when the thickness of the CoNi layer is less than 2 nm. This fact indicates that the current experimental accuracy can fully characterize the SMR effect predicted by the theory. Further estimates based on our experimental accuracy suggest that the order of magnitude of SMR in our system should be less than  $1 \times 10^{-4}$ . This value is far less than the values of SMR in metallic systems with Pt = 10 nm that have been reported by others [5,13]. In addition, the magnitude of AIMR is 4 times larger than that of the theoretical SMR. Therefore, even if SMR appears, the magnetoresistance in the  $yz$  plane is dominated by the AIMR contribution.

## V. RESULTS AND DISCUSSION

The results of the ST-FMR experiment indicate the existence of the spin-orbit torque (SOT). However, such an absorption of spin currents was not evidenced by an apparent SMR. Based on the first model proposed by Chen *et al.* [1], the spin current density  $\mathbf{j}_s^{(\text{F})}$  at the Pt/YIG ( $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ) interface is

$$e\mathbf{j}_s^{(\text{F})}(\mathbf{M}) = G_r\mathbf{M} \times (\mathbf{M} \times \boldsymbol{\mu}_s) + G_i(\mathbf{M} \times \boldsymbol{\mu}_s), \quad (10)$$

where  $G_r(G_i)$  is the real(imaginary) component of the spin-mixing conductance, and  $\boldsymbol{\mu}_s$  is the accumulation of spin at the interface. The spin currents absorbed by YIG via SOT and the reflected spin currents are uniquely influenced by the direction of magnetization, which eventually leads to the SMR effect. Based on this model, the SMR and SOT should happen simultaneously. In the following, we try to figure out such a puzzle from the viewpoint of spin transportation in magnetic heterostructures.

Starting from the interface effect, we first consider the spin memory loss (SML) on the spin transport of the Pt/Co interface [47,48]. SML implies that the spin currents flowing into the CoNi layer are significantly lower than that flowing into the Pt/Co interface in Pt. This type of interfacial absorption of the spin current is independent of the  $\mathbf{M}$  of the CoNi layer, which may decrease the value of SMR. Nevertheless, SML is equivalent to inserting a spin sink layer (SSL) with a specific thickness and spin-diffusion length between the FM and the HM. The spin current at the interface of SSL/FM is similar to Eq. (10), and the Pt layer and SSL can be combined to form an equivalent HM with a smaller  $\theta_{\text{SH}}$ . As the spin current acting on  $\mathbf{M}$  in the FM can be detected using ST-FMR, a lower  $\theta_{\text{SH}} = 0.07$  than previously reports [49] is consistent with the above discussion. However, the magnitude of SMR estimated using  $\theta_{\text{SH}} = 0.07$  is still within the range of our experimental accuracy. Therefore, only considering the SML does not reasonably explain our experimental results.

As SMR was first confirmed and systematically studied in HM/YIG, we believe that the SMR effect in HM/FMI and HM/FM is significantly different due to the different carriers

of spin current and the corresponding scattering mechanisms in FMI and FMs [50–52]. In general, spin current can only be absorbed by FMI through the spin angular-momentum exchange between the localized magnetization  $\mathbf{M}$  in FMI and the conduction-electron spin polarization  $\boldsymbol{\sigma}$  in HMs, i.e., via SOT. However, in FMs, the spin relaxation of itinerant electrons needs to be considered. In 2011, Berger extended Elliott's theory of spin relaxation in normal metals and semiconductors to include metallic ferromagnets [53]. He found that the spin relaxation time is inversely proportional to the spin down resistivity for materials whose spin-up fermi levels are located above the top of the 3d band, such as Ni and Co. Further experimental results confirmed this theory: for instance, Sagasta *et al.* reported that the Elliott-Yafet mechanism is the dominant spin-relaxation mechanism in permalloys [54]; Zhang *et al.* introduced a spin-flip relaxation time to describe the spin transport in FM, and proposed an explicit expression for unidirectional SMR in HM/FM bilayers [55].

With regard to the absence of SMR observed during our experiment, we consider two mechanisms that absorbed the injected spin currents in FMs: SOT ( $\mathbf{M}$  dependent, interfacial effect) and spin relaxation (less  $\mathbf{M}$  independent, bulk effect). The absorption of spin current using SOT is dominant when  $\mathbf{M}$  is perpendicular to  $\boldsymbol{\sigma}$ . Contrarily, when  $\mathbf{M}$  is parallel to  $\boldsymbol{\sigma}$ , SOT is not applicable, but most of the spin currents would be relaxed in FM according to the mechanism proposed by Berger [53]. In general, a majority of the spin currents flowing into FM will be absorbed, resulting in the spin current at the interface that is independent of (or less dependent on) the direction of magnetization. Therefore, although SOT in the Pt/CoNi system was evident, a corresponding magnitude of SMR does not appear. In conclusion, we believe that when considering addition mechanisms for the spin current absorption in FMs, the expression of the spin current at the HM/FMI interface is inapplicable to the HM/FM interface. As a result, SMR performance would be significantly different.

## VI. CONCLUSIONS

In summary, we systematically investigated the magnetoresistance effect in a Pt/(CoNi) $_n$  system with strong PMA and (111) texture. By comparing the magnetoresistance characteristics of the  $yz$  and  $xy$  planes, we determined that the magnetoresistance with interface characteristics in the  $yz$  plane are primarily attributed to AIMR, and no obvious contribution of SMR was found in the experimental results. We obtained the  $\theta_{\text{SH}}$  of Pt through ST-FMR, and the magnitude predicted by the SMR theory was higher than our experimental accuracy. As a consequence, the appearance of  $\cos^2\beta$  terms in the plane perpendicular to the current cannot be used as a fingerprint for the presence of SMR. The separation of the magnetoresistance in the  $yz$  plane introduced by the symmetry breaking of the interface is a necessary prerequisite for the study of the SMR effect in metallic magnetic heterostructures. Moreover, the itinerant magnetism and the corresponding spin-relaxation mechanisms in FMs would result in the spin currents at HM/FM interface that are less dependent on the magnetization, which is responsible for the absence of SMR in our results.

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