Antiferromagnetic interlayer coupling in Pt/Co multilayers with perpendicular anisotropy

Z. Y. Liu,* F. Zhang, H. L. Chen, B. Xu, D. L. Yu, J. L. He, and Y. J. Tian
State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, Qinhuangdao 066004, China
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By using one ultrathin NiO layer to replace the Pt capping layer in the Co/Pt multilayer with perpendicular anisotropy, we present direct evidence for the existence of antiferromagnetic interlayer coupling between the Co layers through the Pt layers thicker than 24 Å. The NiO capping layers play two key roles in the observation of antiferromagnetic interlayer coupling. One is the manipulation of the coercivity of the Co layer in contact with the NiO layer, the other is the enhancement of the electron reflectivities at the Co/Pt interfaces via the specular scattering effect at the Co-NiO interface. The thermal variation in the magnetization of the Co layer also plays an important role in the observed antiferromagnetic coupling due to its strong effect on the phase of electron reflectivity at the Co/Pt interfaces. The magnetization-dependent phase can lead to the ferromagnetic-antiferromagnetic transition of the interlayer coupling at high temperature. All these factors are combined together to realize the direct observation of antiferromagnetic interlayer coupling in the Co/Pt multilayer with perpendicular anisotropy.

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I. INTRODUCTION

Since the discovery of the antiferromagnetic (AF) interlayer exchange coupling (IEC) (Ref. 1) and giant magnetoresistance (GMR) (Ref. 2) in the layered ferromagnetic (FM) films separated by the metallic Cr spacers, these phenomena have been subjects of great interest. A variety of nonmagnetic spacer materials have been investigated and the AF IEC has been observed, for example, in the transition metals.[3,4] To date, most of the layered FM films with the AF IEC have the in-plane anisotropy. In recent years, the observations of the AF IEC in the layered FM films with the perpendicular anisotropy[5–7] have stimulated intensive investigations on these artificial antiferromagnets.[8–14] For their design, the Pt/Co multilayers have been often taken as the FM building blocks. However, the IEC between the Co layers via the Pt layers has been an intriguing problem in the Pt/Co multilayers themselves. For the transition-metallic Pt spacers, although the theoretical calculations predict the existence of the AF IEC,[15] the previous experimental studies have revealed only the FM IEC with the oscillatory coupling strength as a function of the Pt layer thickness resulting from the RKKY-type (Ruderman-Kittel-Kasuya-Yosida) interactions.[16,17] The polarization of the Pt layers owing to the Co 3*d*-Pt 5*d* hybridization at the Pt/Co interfaces, which makes the Pt layers become weakly FM, is considered to play an important role in the FM IEC.[17] Understanding the IEC in the Pt/Co multilayers is significant for the design of the artificial antiferromagnets with perpendicular anisotropy[14] and their applications in spintronic devices.[18]

In the present work, our experimental investigations on the $[\mathrm{Pt/Co}]_3$ multilayers capped by one ultrathin NiO layer have presented unambiguous evidence for the existence of AF IEC between the Co layers across the Pt layers with thickness larger than 24 Å. This AF IEC is found to be thermally dependent, and is very sensitive to the thicknesses of the NiO capping layer and the Pt layer.

II. SAMPLE PREPARATION AND MEASUREMENTS

The samples with the configuration of $[\mathrm{Pt}(t_{\mathrm{Pt}}\ \text{Å})/\mathrm{Co}(4\ \text{Å})]_3/\mathrm{NiO}(t_{\mathrm{NiO}}\ \text{Å})$ were prepared onto the glass substrates covered with 100 Å Pt buffer layer using the dc and rf magnetron sputtering in 4 mTorr Ar pressure. The base pressure before deposition was 4×10^{-7} Torr. The deposition rates from the Pt (99.95%), Co (99.9%), and NiO (98%) targets were calibrated to be 0.28. 0.18, and 0.21 Å/s, respectively. X-ray diffraction measurements display that the samples are highly fcc(111) textured, and the grazing incident x-ray reflectivity confirms the high quality of the interfaces. The measurements of extraordinary Hall effect (EHE) and magnetoresistance (MR) were performed in PPMS (Quantum Design) with the magnetic field and the current applied out of plane and in plane, respectively.

III. RESULTS AND DISCUSSION

For the $[\mathrm{Pt}(t_{\mathrm{Pt}})/\mathrm{Co}(4\ \text{Å})]_3$ multilayer with the NiO capping layer and the Pt layer thickness less than 24 Å, its loop always presents a square shape as shown by inset of Fig. 1. Considering the field precision of 0.1 Oe in PPMS, it can be seen from Fig. 1 that the variation in H_c with the Pt layer thickness of t_{Pt} presents the oscillatory behavior superimposed onto an average decay with the largest oscillatory amplitude reaching up to more than 100 Oe. This observed oscillatory behavior of H_c as a function of t_{Pt} is similar to the previous observations in the pure Co/Pt multilayers.[16,17] The only difference is that the presence of one NiO capping layer leads to the enhancement of the coercivity. The coercivity of H_c is considered to represent the strength of FM IEC between the Co layers across the Pt layers, and its oscillatory dependence on the Pt layer thickness is considered to come from the RKKY-type coupling.[17]

However, with the increase in the Pt layer thickness above 24 Å, we found that in presence of the NiO capping layer, the EHE loop becomes very sensitive to the Pt layer thickness and is no longer in a square shape. Figure 2(a) shows the EHE loop at 300 K for the $[\mathrm{Pt}(26.5\ \text{Å})/\mathrm{Co}(4\ \text{Å})]_3/\mathrm{NiO}(11\ \text{Å})$ multilayer. Clearly, three separate sharp magnetization reversals are observed with two steps formed between them, demonstrating the

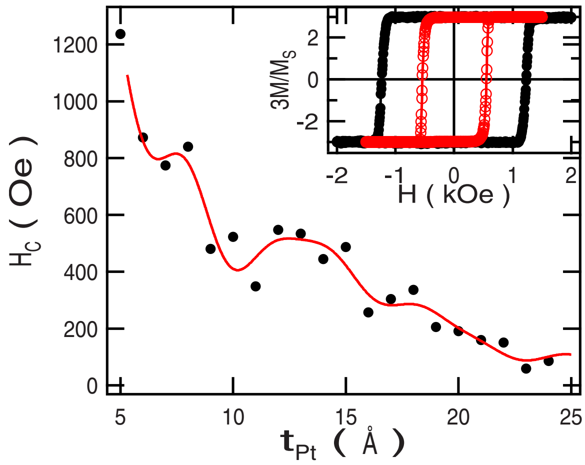

FIG. 1. (Color online) The coercivity of H_c at 300 K of the $[Pt(t_{Pt}\ Å)/Co(4\ Å)]_3/NiO(20\ Å)$ multilayer with $t_{Pt} \leq 24$ Å. The solid line is a guide for the eyes. Inset shows the square EHE loops of the $[Pt(5\ Å)/Co(4\ Å)]_3$ multilayers capped by the 20 Å NiO (solid circle) and Pt (empty circle) layers, respectively. By replacing the Pt capping layer with a NiO layer, the EHE loop is observed to present the square shape but with an enhanced coercivity compared with the Pt capping layer for $t_{Pt} \leq 24$ Å. As a function of the Pt layer thickness of $t_{Pt} \leq 24$ Å, H_c demonstrates the oscillatory behavior superimposed onto an average decay.

separate reversals of the three Co layers. The occurrence of the first sharp reversal in a positive field is strongly suggestive of the existence of AF IEC between the Co layers. Just from the EHE loop is it not possible to determine unambiguously the reversal order in the three Co layers. On the first step after the first sharp transition in the EHE loop, the magnetizations of the three Co layers from top to bottom can have one of the alignments: down-up-up, up-down-up, and up-up-down. In layered magnetic thin-film systems, the GMR effect resides in the spin-dependent scattering, which depends on the relative magnetization directions in the magnetic layers. Figure 2(b) gives the MR curve at 300 K. Along the field's decreasing and increasing branches, the sharp increase to the maximum plateau corresponds to the first sharp switching to the first step in the EHE loop. In view of the spin-dependent scattering, the magnetization alignments of down-up-up and up-up-down cannot make a big difference in the MR value, while the alignment of up-down-up represents the antiparallel alignment of magnetizations, thus giving rise to the maximum plateau of $\Delta R/R$. Hence, among the three Co layers, the first reversal occurs in the middle Co layer. After the maximum plateau, the MR value of $\Delta R/R$ presents a sharp drop to the lower plateau corresponding to the second sharp switching to the second step in the EHE loop. On the second step, the magnetizations in the three Co layers could align either down-down-up or up-down-down. The absolute determination can make use of the exchange biasing effect. If the magnetization of the top Co layer in contact with the NiO capping layer keeps pointing up (down) during cooling down from 300 K to low temperature, the exchange biasing field of the top Co layer will be observed to be negative (positive).[19] We performed the cooling down from 300 to 10 K in one field of −20 Oe on the second step. Figure

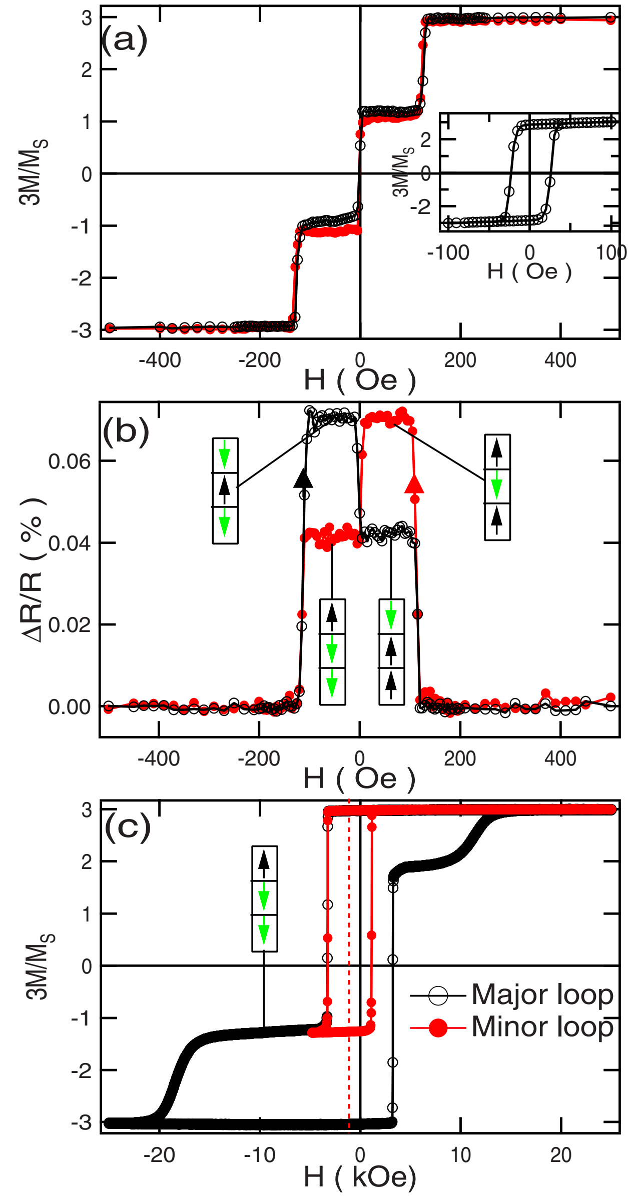

FIG. 2. (Color online) (a) The EHE loop at 300 K for the $[Pt(t_{Pt}\ Å)/Co(4\ Å)]_3/NiO(11\ Å)$ multilayer with t_{Pt}=26.5 Å, displaying three sharp transitions with the almost equal heights. Inset shows the EHE square loop for the same multilayer but with a Pt capping layer of 26.5 Å, being indicative of the simultaneous reversal of the three Co layers. (b) The MR curve at 300 K for the same multilayer. $\Delta R/R$ is defined as $\Delta R/R=[R(H)-R(H_S)]/R(H_S)$, where $R(H)$ and $R(H_S)$ are the measured resistances in the field H and the saturation field H_S, respectively. The alignment of arrows labels out the magnetization orientations of the three Co layers from top to bottom on the step. (c) The EHE major and minor loops at 10 K obtained after the cooling down from 300 to 10 K in the field of −20 Oe chosen on the second step of the EHE loop at 300 K.

2(c) shows the major and minor EHE loops at 10 K after the cooling. A big negative exchange biasing field of −3637 Oe was observed for the top Co layer. Thus, we can conclude that on the second step in Fig. 2(a), the magnetizations in the three Co layers have the alignment of up-down-down, and the top Co layer gives rise to the third sharp transition in the EHE loop and the corresponding sharp decrease in the MR value after the lower plateau. The observed order of magnetization reversals or the different switching fields in the three Co layers could be understood by minimization of the free energy including the magnetocrystalline energy, the exchange coupling at the NiO-Co interface, the interlayer exchange coupling between the Co layers, the Zeeman energy, etc.[20–23]

With the Pt layer thickness larger than 24 Å, the reversal order in the three Co layers is found to be tunable by adjusting the thicknesses of the Pt and NiO layers. Figure 3 gives the typical MR curves at 300 K for the [Pt(t_{Pt} Å)/Co(4 Å)]$_3$/NiO(20 Å) multilayers with t_{Pt} =26.5, 28, 31, 33 Å. Similar to the MR curve shown in Fig. 2(b), separate sharp reversals with steps formed between them are observed along the field's decreasing and increasing branches. The order of magnetization reversals in the three Co layers is determined as shown in Fig. 3. It is found that with increasing the Pt layer thickness above 24 Å, the top and middle Co layers generate the first and last magnetization reversals alternatively. This dependence of the reversal order in the three Co layers indicates the oscillatory behavior of the AF IEC as a function of the Pt layer thickness above 24 Å. Choosing the Pt layer thickness of 26.5 Å, Fig. 2(b) and Figs. 4(a)–4(c) give the MR curves for the NiO capping layers with thicknesses of 11, 14, 16, and 17 Å, respectively. Clearly, similar to the dependence on the Pt layer thickness, for the selected Pt layer thickness of 26.5 Å, with increasing the thickness of the NiO capping layer, the first and last magnetization reversals occur in the top and middle Co layers alternatively, demonstrating the crucial effect of the thickness of NiO capping layer on the AF IEC and the magnetization reversals.

In the pure Pt/Co multilayers with perpendicular anisotropy, the magnetostatic dipolar coupling through the thick Pt layers becomes negligible owing to its quick decay with increasing the Pt layer thickness,[16] and only the weak RKKY-type coupling is observable.[17] In the present work, in the [Pt(t_{Pt})/Co(4 Å)]$_3$ multilayer capped with one ultrathin NiO layer, one of the key roles of the NiO capping layer is to manipulate the coercivities of the Co layers. In bulk NiO, the AF spins align parallel in each (111) plane with the antiparallel alignment in adjacent (111) planes, generating the AF order. For the (111) textured ultrathin NiO capping layer, the AF anisotropy becomes weak so that the AF spins are tilted toward the out-of-plane direction by the direct AF-FM exchange coupling at the Co-NiO interface, leading to the appearance of out-of-plane AF spin components.[10,24–27] The existence of out-of-plane AF spin components is the key for the out-of-plane exchange bias. However, for the ultrathin NiO layer, its blocking temperature has been shown to be much lower than room temperature.[24,25] Above the blocking temperature, the AF anisotropy becomes too weak to fix the AF spins in position; in other words, the AF spins are no longer frozen to create the exchange bias or a shift of the hysteresis loop. Owing to the direct AF-FM spin coupling at the Co-NiO interface, the unfrozen AF spins are rotatable with the FM spins. The AF rotatable spins manipulate the coercivity between the AF layer and the FM one,[28] enhancing the FM

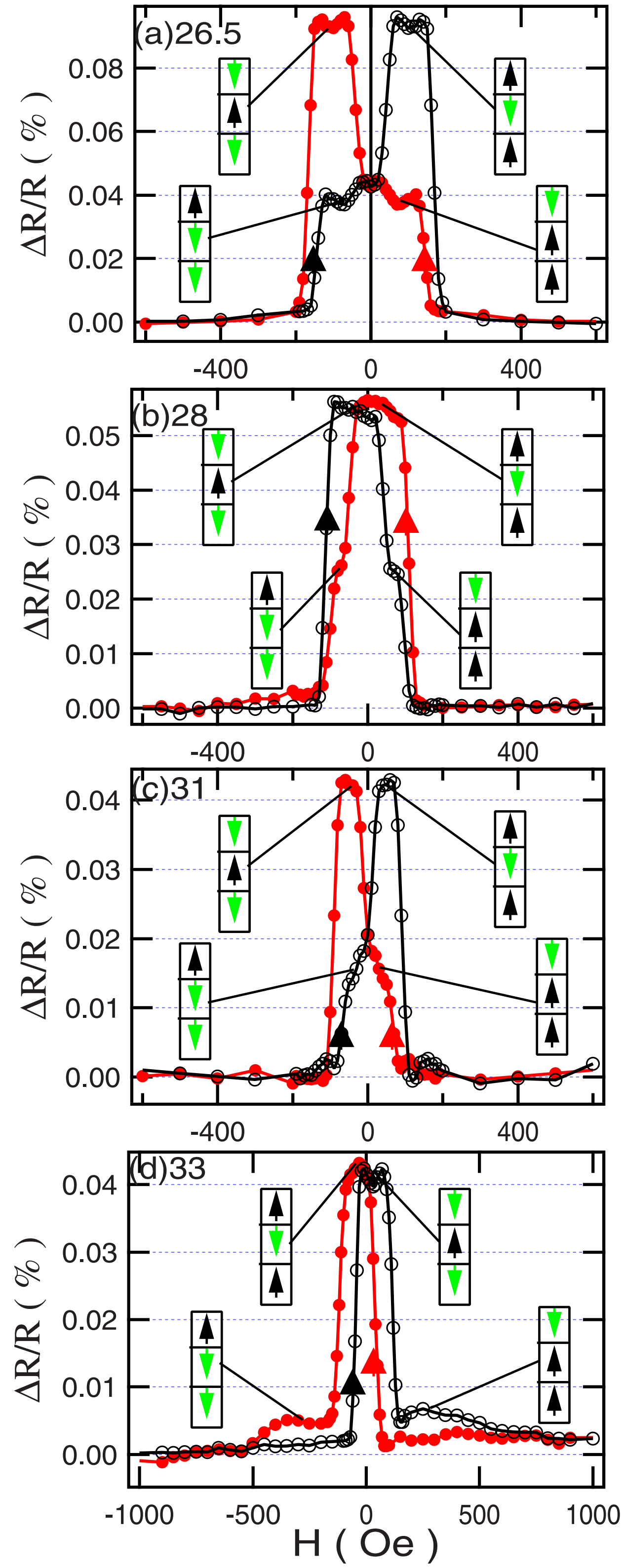

FIG. 3. (Color online) The MR curves at 300 K for the [Pt(t_{Pt} Å)/Co(4 Å)]$_3$/NiO(20 Å) multilayers with (a) t_{Pt} =26.5 Å, (b) 28 Å, (c) 31 Å, and (d) 33 Å. The alignment of arrows labels out the magnetization directions on the step in the three Co layers from top to bottom. Clearly, with increasing the Pt layer thickness above 24 Å, the first and third magnetization reversals occur in the top and middle Co layers alternatively.

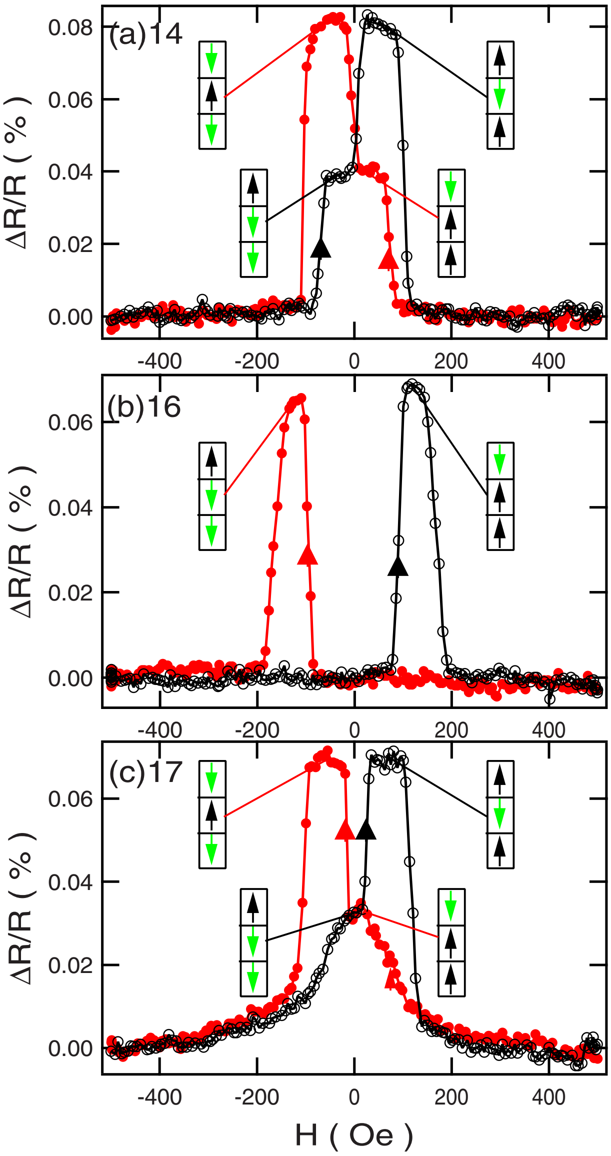

FIG. 4. (Color online) The MR curves at 300 K for the [Pt(26.5 Å)/Co(4 Å)]$_3$/NiO(t_{NiO} Å) multilayers with (a) t_{NiO} $=14$ Å, (b) 16 Å, and (c) 17 Å. The alignment of arrows shows the magnetization directions on the step in the three Co layers from top to bottom. Clearly, with increasing the thickness of NiO capping layer, the top and middle Co layers give rise to the first and last magnetization reversals alternatively.

coercivity. In the present case, the top Co layer in contact with the NiO capping layer is separated from the other two Co layers by a thick Pt layer of 26.5 Å. The direct AF-FM spin coupling at the Co-NiO interface affects mainly the coercivity of the top Co layer but has negligible effect on the middle and bottom Co layers, which is similar to the case of a spin-valve structure. Thus, the coercivity of the top Co layer can be different from those of the other two Co layers, making it easy for the direct observation of AF interlayer coupling between the Co layers. The dominant effect of the direct AF-FM spin coupling at the Co-NiO interface on the top Co layer is further confirmed by the measurements of the exchange biasing effect after the field cooling from 300 to 10 K in one field of −20 Oe. As shown in Fig. 2(c), at 10 K, the middle and bottom Co layers switch together with the symmetric switching fields along the field's decreasing and increasing branches with no shift observed along the field direction, and their minor-loop center shows a big shift of 1092 Oe along the negative field direction; while for the top Co layer, a big negative biasing field of −3637 Oe is observed. These phenomena tell the following information: (1) the AF-FM spin coupling at the Co-NiO interface affects mainly the top Co layer for the thick Pt layer; (2) at low temperature, the IEC between the Co layers becomes FM. One more key role of the NiO capping layer comes from the specular electron scattering effect at the Co-NiO interface. With one ultrathin NiO layer replacing the Pt capping layer, the Co-NiO interface has the higher potential barrier, at which the specular electron scattering can occur.[29] In spite of the small thickness of the NiO capping layer, its Néel temperature can reach the room temperature or even above,[30] and the higher potential at the Co-NiO interface can be spin dependent owing to the interfacial Ni magnetic moments on the AF NiO layer.[4] When the conduction electrons travel to the Co-NiO interface, they can be reflected back into the Pt/Co multilayer, increasing their mean free path and thus enhancing the spin-dependent electron reflectivities at the Pt/Co interfaces.

During warming up after the field cooling, the measured major and minor loops demonstrate a transition from the FM to AF IEC at 210 K, above which the AF IEC becomes observable, indicating that the AF IEC is thermally dependent. At room temperature, the Pt polarization occurs in about one atomic layer,[31] but it goes deeply into the Pt layer with decreasing temperature, strengthening the RKKY-type FM coupling.[17] Thus, the enhanced Pt polarization favors the occurrence of the FM IEC at low temperature. With increasing the temperature, the transition of FM to AF IEC or the sign reversal of the IEC can be attributed to the following reasons: (1) The thermal variations in the magnetizations of the Co layers; (2) the enhanced multiple electron reflections at the Pt/Co interfaces. A recent study on the sign reversal of the IEC in Gd/Y/Tb trilayers[32] reported that the variations in the magnetizations of the FM layers have strong effects on amplitudes and phases of the spin-dependent electron reflectivities at the interfaces, leading to the magnetization-dependent phase of electron reflectivity as $\phi \approx \pi m(T)$. This magnetization-dependent phase induces the temperature-dependent sign reversal of the IEC. Our measurements on one 4 Å Co layer sandwiched between two Pt layers (not shown here) demonstrate that its magnetization has a temperature dependence of $M_S(T)=M_S(0)(1-T/404)^{0.34}$ with the determined Curie temperature of 404 K. At high temperature close to 300 K, the magnetizations of the Co layers have a strong temperature dependence, which can affect strongly the amplitudes and phases of the spin-dependent electron reflectivities at the Pt/Co interfaces and induce the occurrence of the AF IEC. According to the quantum well model of Stiles,[15] if the Pt spacer is taken to separate the FM layers, the IEC will oscillate as a function of the Pt spacer thickness in multiple periods, among which the strongest oscillation is expected to have a period of about 6 Å. The obtained expression of the IEC contains one oscillatory term represented by a sine function of $\sin(2\pi d/\lambda+\phi)$, where d is the thick-

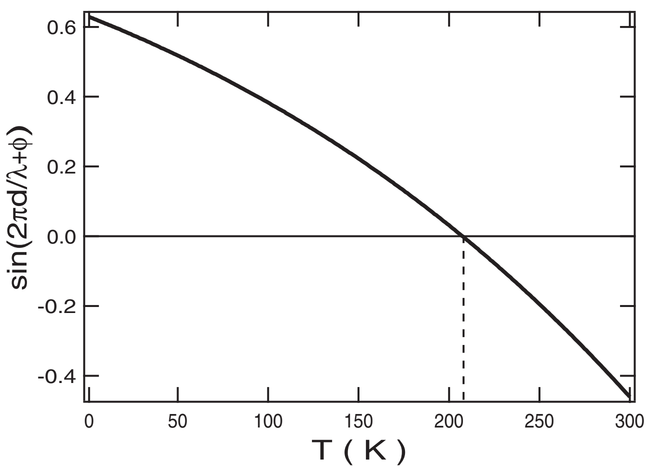

FIG. 5. The calculated expression of $\sin(2\pi d/\lambda+\phi)$ as a function of temperature. In the calculation, the thickness d of the Pt layer is taken to be 26.5 Å, the oscillation period λ is taken to be 5.75 Å, and the magnetization-dependent phase ϕ of electron reflectivity at the Co/Pt interfaces is represented by the expression of $\phi=\pi m(T)$ obtained by Döbrich *et al.* (Ref. 32). $m(T)$ is the normalized magnetization of one 4 Å Co single layer sandwiched between two Pt layers, which is determined to follow the expression of $m(T)=(1-T/404)^{0.34}$. Clearly, with the increase in temperature, the magnetization-dependent phase ϕ is able to induce the sign reversal of $\sin(2\pi d/\lambda+\phi)$, in other words, the FM-AF transition of IEC.

ness of Pt spacer, λ is the oscillatory period, and ϕ is the phase of electron reflectivity at the interface. In our investigated sample of $[\mathrm{Pt}(t_{\mathrm{Pt}}\ \text{Å})/\mathrm{Co}(4\ \text{Å})]_3/\mathrm{NiO}$, if the Pt layers are thick enough, for the three Co layers, the temperature dependence of their magnetizations could be approximately represented by the expression of $m(T)=M(T)/M_S(0)=(1-T/404)^{0.34}$ obtained for one single Co layer sandwiched between two Pt layers. Assuming that the magnetization-dependent phase of the electron reflectivity at the Co/Pt interfaces is represented by the expression of $\phi\approx\pi m(T)$ obtained by Döbrich *et al.*,[32] we calculated the expression of $\sin(2\pi d/\lambda+\phi)$ as a function of temperature (see Fig. 5). In the calculations, the Pt layer thickness of d was taken to be 26.5 Å, and λ is taken to be around 6 Å, the period of the strongest oscillation obtained for the (111) Pt spacer in the calculations by Stiles.[15] As shown in Fig. 5, the magnetization-dependent phase of $\phi\approx\pi m(T)$ is able to induce the sign reversal of $\sin(2\pi d/\lambda+\phi)$, generating the FM-AF transition of the IEC with the increase in temperature. Hence, the observed AF IEC at high temperature is thermally induced.

The previous experimental results suggest that the multiple scattering of electron waves acting in phase is able to strengthen the IEC.[4] The expected AF IEC in the Pt/Co multilayer can be enhanced and become strong enough to suppress the weak FM coupling preferred by the Pt polarization, making it observable in experiments. Moreover, due to the main effect of the NiO capping layer on the top Co layer, its coercivity becomes different from those of the middle and bottom Co layers. Thus the $[\mathrm{Pt}(t_{\mathrm{Pt}})/\mathrm{Co}(4\ \text{Å})]_3$ multilayer capped by the NiO layer behaves similar to a spin-valve structure, making it easy for the experimental observation of the AF IEC. The oscillatory behavior of the AF IEC as a function of the Pt layer thickness is suggestive of the intrinsic RKKY-type nature. The oscillatory behavior of the AF IEC with the increase in the NiO capping layer thickness may be related to the improved AF ordering in the NiO layer.

In summary, our investigations present direct evidence for the existence of the AF IEC in the Pt/Co multilayers with perpendicular anisotropy via one ultrathin NiO capping layer. The ultrathin NiO capping layer plays two key roles in the observation of AF IEC in the Co/Pt multilayer. One role is to manipulate the coercivities of the Co layers, making the coercivity of the top layer quite different from those of the middle and bottom Co layers. The other role is the specular electron reflectivity at the Co-NiO interface, which enhances the traveling distances of electrons in the Co/Pt multilayer. Moreover, the thermal variation in the magnetizations of the Co layers also plays an important role in the observation of AF IEC in the Co/Pt multilayer. The thermal variation in the Co magnetization has a great effect on the phases of the spin-dependent electron reflectivities at the Co/Pt interfaces, leading to the high temperature dependence of the phases. At high temperature, the temperature-dependent phases are able to induce the FM-AF transition of IEC. Combination of these factors makes it successful to observe the AF IEC in the Co/Pt multilayer. Owing to these phenomena, the Pt/Co multilayers could find practical applications in temperature-sensitive devices via the sign reversal of IEC, which can be tuned by properly selecting the Pt layer thickness and the oxide capping layer.

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*liuzy0319@yahoo.com; fhcl@ysu.edu.cn

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