Oscillatory antiferromagnetic interlayer coupling in Co(4 Å)/Pt(t_{Pt} Å)/[Co(4 Å)/Pt(6 Å)/Co(4 Å)]/NiO(20 Å) multilayers with perpendicular anisotropy

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In this work, we have demonstrated the existence of oscillatory antiferromagnetic interlayer coupling in the Co/Pt multilayer with perpendicular anisotropy by using the sample configuration of [Pt(6 Å)/Co(4 Å)]/Pt(t_{Pt} Å)/[Co(4 Å)/Pt(6 Å)/Co(4 Å)]/NiO(20 Å). As a function of t_{Pt}, there exists a transition of the interlayer coupling from ferromagnetic to antiferromagnetic. At $t_{Pt}<25$ Å, the interlayer coupling between Co layers is always ferromagnetic, while at $t_{Pt}>25$ Å, the antiferromagnetic interlayer coupling appears and its strength varies as a function of t_{Pt} in such a way as to lend support to a theoretical model for a (111) Pt spacer.

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Since the first discovery of antiferromagnetic (AF) interlayer coupling in layered magnetic thin film system composed of ferromagnetic layers with in-plane anisotropy separated by metallic spacers,[1] especially the later observation of giant magnetoresistance[2] in antiferromagnetically coupled layered magnetic thin film systems and its important applications in electronic devices and magnetic recording technology,[3–5] extensive investigations have been performed on the interlayer coupling across the nonmagnetic transition-metallic spacers. For most transition-metallic spacers, antiferromagnetic interlayer coupling has been observed, and the coupling strength has been found to increase exponentially with the increase of *d* electron number. One phenomenon common to the interlayer coupling across the nonmagnetic transition-metallic spacers is that as a function of the spacer thickness, the coupling oscillates between ferromagnetic and AF with the periods determined by the geometry of the spacer Fermi surface.[6–8] Surprisingly, for the transition-metallic Pt with electron configuration of $5d^9$, although theoretical calculations[8] have predicted oscillatory AF interlayer coupling with multiperiods for (111) textured Pt spacers, experimental studies[7,9] have given inconsistent results. The studies on Co/Pt multilayers by Parkin demonstrated no existence of AF coupling,[7] while the studies by Le Dang *et al.* reported the existence of AF interlayer coupling but did not presented any evidence of oscillations.[9]

So far, almost all the investigations of the interlayer coupling have been carried out on layered magnetic thin film systems with in-plane magnetization with only a few studies reported on the systems with out-of-plane anisotropy. Specifically, for Co/Pt multilayers with very thin Co layers, their perpendicular anisotropy has attracted considerable attention because of its potential applications in perpendicular magnetic recording technology. It has long been considered that the ultrathin Co layers are ferromagnetically coupled together across the Pt layers.[10–12] Recently, AF interlayer coupling has been claimed to be observed in the measurement of virgin magnetization curve of the perpendicular spin valve composed of Co/Pt multilayers, but the measurement of hysteresis loop demonstrated only a ferromagnetic (FM) interlayer coupling.[11,12] In our recent studies on the interlayer coupling in the Co/Pt multilayers with perpendicular anisotropy, very weak noncollinear interlayer coupling has been really observed.[13] However, to date, no direct experimental evidence has been found to show the existence of oscillatory AF interlayer coupling across the Pt layers in Co/Pt multilayers with perpendicular anisotropy. In the Co/Pt multilayers with perpendicular anisotropy, especially when the Pt layers are not thick and the number of Co layers is big, the interfacial roughness, existence of pinholes in the Pt layers, and defects formed during preparation are able to induce the long-range magnetostatic interactions between the Co layers. The ferromagnetic interlayer coupling originating from the magnetostatic interactions could be strong enough to suppress the weak AF interlayer coupling if it exists, making the observation of AF interlayer coupling across the Pt layers very difficult.

In this work, in order to explore the possible existence of AF interlayer coupling in the Co/Pt multilayer with perpendicular anisotropy, we have tried to lower the magnetostatic interactions by using just three Co layers in the samples and the deposition rate of Pt as low as possible. The samples have been designed with spin-valve configuration of Co(4 Å)/Pt(t_{Pt} Å)/[Co(4 Å)/Pt(6 Å)/Co(4 Å)]/NiO(20 Å) by making use of our previous observation of enhanced coercivity in a Co/Pt multilayer capped with an ultrathin NiO layer.[14] For an ultrathin NiO film, its Néel temperature is generally much lower than the bulk value of 520 K, but it can still reach up to room temperature (RT) or even higher.[15] When it is in contact with a FM film, its blocking temperature for the appearance of any loop shift is much lower than RT. In this structure, the two Co layers in [Co(4 Å)/Pt(6 Å)/Co(4 Å)] are ferromagnetically coupled together due to the thin separating Pt layer. Owing to the sample preparation in the absence of field, the significant domain structure formed in the AF NiO capping layer leads to the greatly enhanced coercivity in [Co(4 Å)/Pt(6 Å)/Co(4 Å)], which is much larger than that of the bottom Co single layer (referring to Ref. 14 for a detailed discussion). The Pt spacer thickness was changed

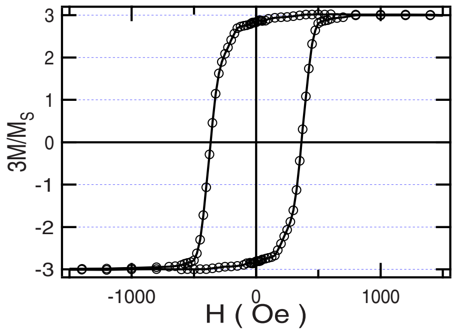

FIG. 1. (Color online) One typical loop at 300 K for $t_{Pt}=25$ Å. Considering that there are three Co layers with the same thickness in all the samples, the magnetization M was normalized to 3 by the saturation magnetization M_S in all the measured loops. The loops at 300 K for $t_{Pt}<25$ Å have the same shape. The almost square shape implies that the bottom Co single layer and [Co(4 Å)/Pt(6 Å)/Co(4 Å)] are ferromagnetically coupled together across the Pt spacer with $t_{Pt}\leq 25$ Å.

from 6 to 70 Å. Between the bottom Co single layer and [Co(4 Å)/Pt(6 Å)/Co(4 Å)] across the Pt spacer of thickness t_{Pt}, if there exists any interlayer coupling, the difference in their switching fields makes it possible to directly determine the coupling nature and obtain the coupling strength J_{iec} via measurement of the minor loop of the bottom Co single layer.

The samples were deposited by magnetron sputtering from Co (99.9%), Pt (99.99%), and NiO (99.5%) targets onto glass substrates covered with 100 Å Pt buffer layer in 4 mTorr Ar pressure. The base pressure before deposition was 4×10^{-7} Torr. The deposition rates of Co, Pt, and NiO were 0.21, 0.28, and 0.23 Å/s, respectively. X-ray diffraction measurements reveal a (111) texture of the samples. The hysteresis loop measurements were performed using a sample vibrating magnetometry attached to the physical property measurements system (PPMS). Magnetoresistance measurements were also performed on some samples in PPMS using the standard four-probe method with current applied in plane. During all the measurements, the magnetic field was always applied perpendicular to the sample surface.

Measurements of hysteresis loops at 300 K have demonstrated a transition of the interlayer coupling from ferromagnetic to AF with increasing Pt spacer thickness t_{Pt}. When Pt spacer thickness t_{Pt} is less than 25 Å, the loop is always in an almost square shape, as shown in Fig. 1, being indicative of the ferromagnetic interlayer coupling between the bottom Co single layer and [Co(4 Å)/Pt(6 Å)/Co(4 Å)]. In the Co/Pt multilayers with perpendicular anisotropy, previous studies have revealed that the ferromagnetic interlayer coupling strength oscillates as a function of the Pt layer.[10] However, when Pt spacer thickness t_{Pt} becomes larger than 25 Å, the loop is no longer in a square shape. Figure 2 gives the magnetization and magnetoresistance (MR) loops for three selected Pt spacer thicknesses of $t_{Pt}>25$ Å. Clearly, along both the field decreasing and increasing branches of the magnetization loop, there appear two sharp transitions with a step forming between them. On the step, the magnetizations of the bottom Co single layer and [Co(4 Å)/Pt(6 Å)/Co(4 Å)] align antiparallel to each other, which can be confirmed by the measurement of spin-dependent scattering MR. The spin-

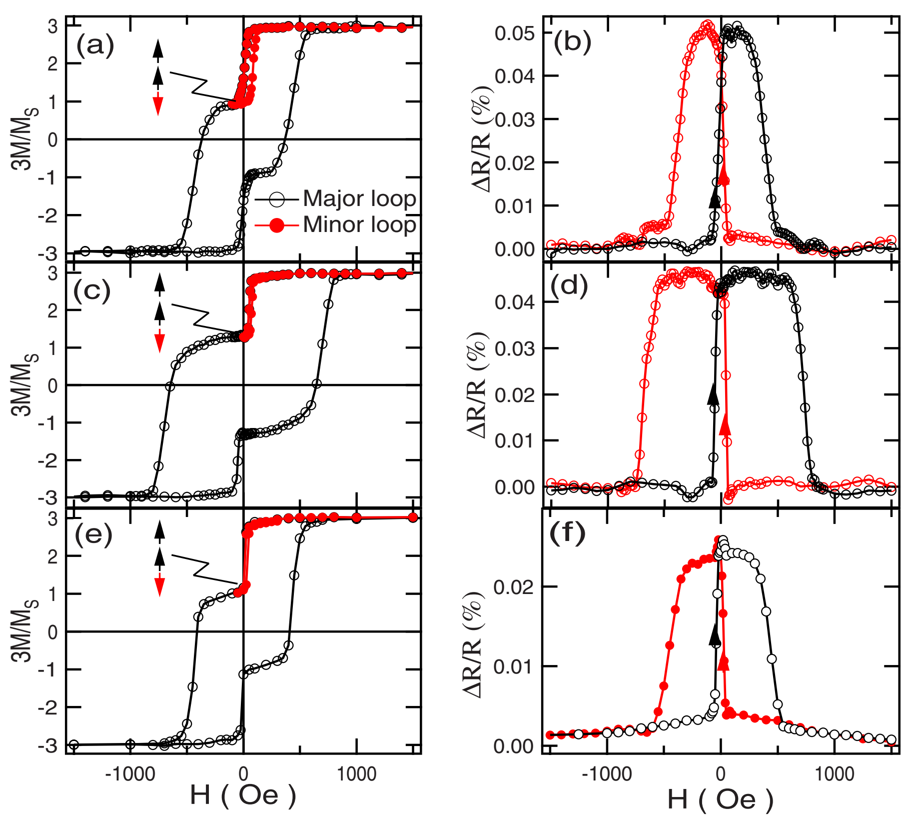

FIG. 2. (Color online) Major and minor magnetization loops and magnetoresistance (MR) loops at 300 K for three selected Pt spacer thicknesses of [(a) and (b)] $t_{Pt}=28$ Å [(c) and (d)] 31 Å, [(e) and (f)] 40 Å. $\Delta R/R$ was defined as $\Delta R/R=[R(H)-R(H_S)]/R(H_S)$, where $R(H)$ and $R(H_S)$ are the MR values taken at 300 K in the field of H and the saturation field of H_S, respectively. At $t_{Pt}>25$ Å, the magnetization loops have the same stepped shape. Arrow alignments are used to represent the magnetization directions in the three Co layers after the first sharp switching. In the MR loops, the sharp increase and decrease in $\Delta R/R$ correspond to the first and second sharp switching in the magnetization loops, and the maximum plateau of $\Delta R/R$ corresponds to the step between the first and second sharp switching.

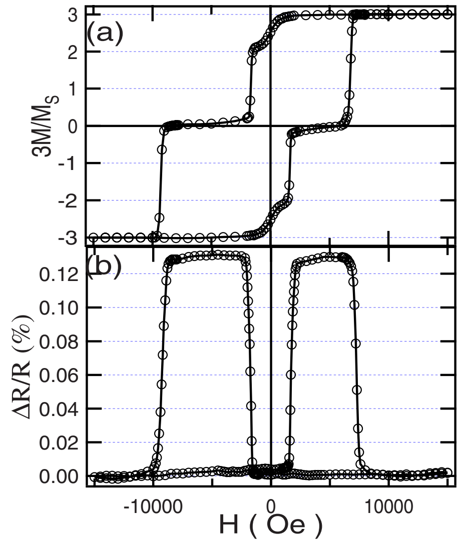

FIG. 3. (Color online) (a) The loop measured at 40 K for $t_{\mathrm{Pt}}=40$ Å after the cooling from 300 to 40 K in zero field right after the first sharp switching at 300 K, as shown in Fig. 2(e). Clearly, the second sharp switching field along the field decreasing branch is shifted greatly toward the negative field direction in comparison with that along the field increasing branch, indicating that in zero field right after the first sharp switching at 300 K, the magnetization of [Co(4 Å)/Pt(6 Å)/Co(4 Å)] points up. (b) The corresponding magnetoresistance loop at 40 K after the cooling for $t_{\mathrm{Pt}}=40$ Å.

dependent scattering MR is dependent on the relative orientation of magnetizations in the magnetic layers, and in the MR loops, as shown Fig. 2, the sharp increase and decrease in $\Delta R/R$ correspond to the first and second sharp switching in the magnetization loop, respectively, and the maximum plateau of $\Delta R/R$ corresponds to the step between the first and second sharp switching. On the step after the first sharp switching in the magnetization loop, the appearance of maximum plateau of $\Delta R/R$ confirms the antiparallel alignment of magnetizations in the bottom Co single layer and the top two ferromagnetically coupled Co layers. In zero field right after the first sharp switching along the field decreasing branch of the loop for $t_{\mathrm{Pt}}=40$ Å [see Fig. 2(e)], field cooling has been performed from 300 to 40 K. As it is well known, NiO is one kind of AF materials. In the loop measured at low temperature, the field-cooling-induced exchange biasing at the Co-NiO interface, which is directly determined by the direction of magnetization in [Co(4 Å)/Pt(6 Å)/Co(4 Å)] at 300 K, is able to produce a big difference in the switching fields of [Co(4 Å)/Pt(6 Å)/Co(4 Å)] along the field decreasing and increasing branches of the loop. As shown in Fig. 3, the measured magnetization and MR loops at 40 K after cooling demonstrate clearly that the second sharp switching originating from [Co(4 Å)/Pt(6 Å)/Co(4 Å)] along the field decreasing branch of loop is shifted greatly toward the negative field direction in comparison with that along the field increasing branch, suggesting strongly that the magnetization of [Co(4 Å)/Pt(6 Å)/Co(4 Å)] keeps pointing up on the step after the first sharp switching at 300 K [see Fig. 2(e)]. Hence, the first switching at 300 K is attributed to the switching down of magnetization in the bottom Co single layer.

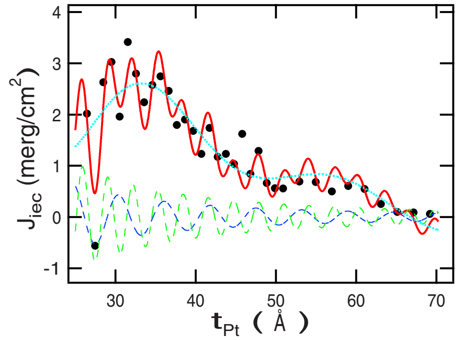

FIG. 4. (Color online) Dependence of AF interlayer coupling strength J_{iec} on the Pt spacer thickness of t_{Pt} at 300 K. The solid dots are experimental data. The solid line is a representation of the experimental data from the expression of $J_{\mathrm{iec}}=p_0+p_1\exp(-p_2t_{\mathrm{Pt}}+p_3)+\Sigma_{i=1}^{3}A_i\sin(k_it_{\mathrm{Pt}}+\varphi_i)/t_{\mathrm{Pt}}^2$ with $(p_0,p_1,p_2,p_3)=(-140.67,533.33,0.0064,-0.89)$, $(A_1,A_2,A_3)=(11\,759.05,7102.60,-14\,127.07)$, $(k_1,k_2,k_3)=(2.0,1.10,0.27)$, and $(\varphi_1,\varphi_2,\varphi_3)=(0.21,-0.59,2.81)$. The dotted line was the addition of the exponential term and the oscillatory term with $k_3=0.27$ and the dashed lines were calculated from the oscillatory terms with $(k_1,k_2)=(2.0,1.10)$, respectively.

Owing to the big difference in switching fields of the bottom Co single layer and [Co(4 Å)/Pt(6 Å)/Co(4 Å)] at 300 K, the nature and strength of the interlayer coupling have been determined directly by the measurement of the minor loop of the bottom Co single layer. As shown in Fig. 2, the obvious positive shift H_{iec} of the minor-loop center is suggestive of the existence of AF interlayer coupling between the bottom Co single layer and [Co(4 Å)/Pt(6 Å)/Co(4 Å)], favoring the antiparallel alignment of magnetizations on the step. The coupling strength is determined directly by the minor-loop shift H_{iec} via $J_{\mathrm{iec}}=H_{\mathrm{iec}}M_St_{\mathrm{Co}}$, where M_S (=1400 emu) and t_{Co} (=4 Å) are Co saturation magnetization and thickness of the Co layer, respectively. The dependence of J_{iec} on Pt spacer thickness t_{Pt} is presented in Fig. 4. AF interlayer coupling can be observed across the Pt spacer with thickness at least up to 70 Å, and the strongest coupling strength is obtained to be 3.42 merg/cm^2, being pretty weak. With the increase of t_{Pt} above 25 Å, the coupling strength J_{iec} varies in such a way as to indicate a long wavelength (small k) oscillation and, possibly, a short wavelength (large k) period as well.

Following the formulation of Stiles,[8] the energy E_{iec} of oscillatory interlayer coupling across the (111) textured Pt spacer can be represented in a simple expression

$$E_{\mathrm{iec}}=\sum_{i=1}^{3}A_i\sin(k_it_{\mathrm{Pt}}+\varphi_i)/t_{\mathrm{Pt}}^2, \quad (1)$$

where A_i are related to the geometrical weights and differences in the reflection amplitudes for the different Fermi surface configurations of the Pt spacer, φ_i are the phase changes appearing on integration over parallel momentum, and k_i are the Fermi wave vectors determining the periods via $d_i=2\pi/k_i$. According to previous studies on the interlayer coupling in the Co/Pt multilayers with perpendicular anisotropy,[11] the coupling can be considered to be composed of two components: one is the magnetostatic interactions originating from roughness, which decays exponentially with increasing the Pt spacer thickness, and the other one is the Ruderman-Kittel-Kasuya-Yosida (RKKY)-type coupling, giving rise to the oscillations of coupling. In the present work, if the magnetostatic interaction induced by roughness is considered and its variation with t_{Pt} is represented by a simple exponential function of $p_0+p_1\exp(-p_2 t_{\mathrm{Pt}}+p_3)$ (p_0, p_1, and p_3 are three constants), following the previous studies,[11] the interlayer coupling energy in the Co/Pt multilayer is possible to be simply expressed as

$$J_{\mathrm{iec}} = p_0 + p_1 \exp(-p_2 t_{\mathrm{Pt}} + p_3) + \sum_{i=1}^{3} A_i \sin(k_i t_{\mathrm{Pt}} + \varphi_i)/t_{\mathrm{Pt}}^2. \tag{2}$$

The dependence of the AF interlayer coupling strength on t_{Pt} has been represented using Eq. (2). The data are not suitable for fitting to obtain the exact amplitudes of the oscillations, but consistency with the three dominant periods (k_i for $i=1-3$) of 25, 7, and 5 Å predicted by Stiles[8] is obtained. Our least squares minimization yields periods of 23, 6, and 3 Å, respectively.

In summary, we have clearly demonstrated that AF interlayer coupling exists in the Co/Pt multilayer with perpendicular anisotropy. The AF interlayer coupling strength has been found to vary above 25 Å in such a way as to agree, qualitatively, with the three dominant periods predicted by the calculations of Stiles.[8] This discovery suggests that AF interlayer coupling should be considered in investigations on magnetic properties of Co/Pt multilayers with perpendicular anisotropy, especially when the Pt layer is thick enough.

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