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SPECIAL TOPIC — 60th Anniversary of Department of Physics of Nanjing Normal University

Temperature-dependent interlayer exchange coupling strength in synthetic antiferromagnetic $[Pt/Co]_2/Ru/[Co/Pt]_4$ multilayers*

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In this work, we experimentally investigated the thermal stability of the interlayer exchange coupling field (H_{ex}) and strength ($-J_{iec}$) in synthetic antiferromagnetic (SAF) structure of $[Pt(0.6)/Co(0.6)]_2/Ru(t_{Ru})/[Co(0.6)/Pt(0.6)]_4$ multilayers with perpendicular anisotropy. Depending on the thickness of the spacing ruthenium (Ru) layer, the observed interlayer exchange coupling can be either ferromagnetic or antiferromagnetic. The H_{ex} were studied by measuring the magnetization hysteresis loops in the temperature range from 100 K to 700 K as well as the theoretical calculation of the $-J_{iec}$. It is found that the interlayer coupling in the multilayers is very sensitive to the thickness of Ru and temperature. The H_{ex} exhibits either a linear or a non-linear dependence on the temperature for different thickness of Ru. Furthermore, our SAF multilayers show a high thermal stability even up to 600 K ($H_{ex} = 3.19$ kOe, $-J_{iec} = 1.97$ erg/cm^2 for $t_{Ru} = 0.6$ nm, the unit 1 Oe = 79.5775 A·m^{-1}), which was higher than the previous studies.

Keywords: synthetic antiferromagnetic, exchange coupling field, interlayer exchange coupling strength

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1. Introduction

Spin-transfer-torque magnetic random access memories (STT-MRAMs) is one of the most attractive candidate to satisfy the need of memory markets owing to their outstanding performance, such as non-volatile, low power consumption, unlimited endurance, wide operating temperature, and long-term data retention.[1–8] However, how to realize the high density of current MRAM is the main bottleneck limiting their practical applications, which is due to the stray magnetic fields generated by magnetic layers. Recently, perpendicularly magnetized tunnel junctions (p-MTJs) that including synthetic antiferromagnetic (SAF) layers have been used to promote the development of STT-MRAMs. The introduction of SAF layers into MRAM devices suppresses the effect from the stray magnetic field, extends the operation temperature range, and improves the thermal stability.[9–11]

The SAF structure is mainly composed of two ferromagnetic (FM) layers separated by a nonmagnetic spacing layer (*e.g.*, Ru, Rh, or Ir),[12–14] where the two FM layers are antiferromagnetically or ferromagnetically coupled by varying the thickness of the spacing layer. Thanks to their perceived advantages of strong magnetic stability, high thermal stability, and sensitive magnetic moment rotation, SAF structure with perpendicular magnetic anisotropy (PMA) has been extensively studied.[15,16] For instance, the spin-orbit torque induced switching of the two magnetic layers in Co/Ru/$[Co/Pt]_2$[17] and completely compensated Pt/[Co/Pd]/Ru/[Co/Pd][18] SAF structures. The antiferromagnetic interlayer exchange coupling (IEC) and the outstanding performance in a wider temperature range are the key fundamental characteristics of SAF multilayers, which are important for such large-scale industrial application as field sensors, magnetic recording, and magnetic random-access memory.[19,20] As required by the potential military and aerospace applications, it is highly desirable to control the IEC of multilayers by changing external physical parameter, such as temperature. However, the widespread Pt/Co-based SAF multilayer with an Ru spacer is still a challenge due to the structural discontinuity or intermixing that occurs at high temperature. In this work, we experimentally investigated the temperature-dependent IEC properties of SAF multilayers comprised of two Pt/Co ferromagnetic layers and an Ru spacing layer. It is showing that the interlayer exchange coupling field (H_{ex}) and coupling strength ($-J_{iec}$) are highly dependent on the Ru spacer thickness (from 0 nm to 1.6 nm) and the measuring temperature (from 100 K to 700 K).

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2. Experimental method

Ta(4)/Pt(4)/[Pt(0.6)/Co(0.6)]$_2$/Ru(t_{Ru})/[Co(0.6)/Pt(0.6)]$_4$/Ta(4) thin film stacks were deposited on a thermally oxidized silicon wafer use DC magnetron sputtering techniques at room temperature (RT). Argon gas ($\sim$ 2.3 mTorr, 1 Torr = 1.33322×10^2 Pa) was used during the sputtering process with a background pressure of 2×10^{-9} Torr, and the deposition rates for Ta, Pt, Co, and Ru were 0.026, 0.544, 0.14, 0.21, and 0.10 Å/s, respectively. Numbers in parentheses are the nominal thicknesses in unit nm. In the thin film stacks, the bottom ferromagnetic stack ([Pt(0.6)/Co(0.6)]$_2$) and the top ferromagnetic stack ([Co(0.6)/Pt(0.6)]$_4$) are antiferromagnetically or ferromagnetically coupled through the spacing layer depending on the Ru thickness. We prepared five SAF multilayers with the thickness of Ru, $t_{Ru} = 0$, 0.6, 0.9, 1.2, 1.6 nm, respectively. To compare the effect of the thickness of FM layer [Pt(0.6)/Co(0.6)]$_x$, we also prepared two reference samples Ta(4)/Pt(4)/[Pt(0.6)/Co(0.6)]$_x$/Ta(4) ($x = 2$ and 4, respectively). For simplicity, these two reference samples will be named as 2ML and 4ML. Vibrating sample magnetometer (VSM) was used to measure the in-plane and out-of-plane magnetization hysteresis loops of the sputtered magnetic thin film stacks. A series of hysteresis curves were achieved by varying the magnetic field at center sweeping rate using electro-magnet at different temperatures. The measurements done below RT were accomplished by a continuous flow of gas through liquid nitrogen cooled, and the measurements done above RT were carried out with the sample chamber heated electrically.

3. Results

Figure 1(a) shows the representative schematic for the SAF multilayer structure. Figure 1(b) illustrates the out-of-plane magnetic hysteresis (M–H) loop for the SAF multilayers with Ru spacer thickness of $t_{Ru} = 0.6$ nm and $t_{Ru} = 0$ nm at room temperature. Without the presence of the Ru spacer, the $t_{Ru} = 0$ nm SAF multilayer exhibits a ferromagnetic coupling (FC) character, *i.e.*, single-step switching. In contrast, with the presence of the Ru spacer, the $t_{Ru} = 0.6$-nm SAF multilayer exhibits a typical antiferromagnetic coupling (AFC) character, *i.e.*, multi-step switching. The perpendicular interlayer exchange coupling field (H_{ex}) is defined as the field shift of the minor loop as indicated in Fig. 1(b). Figure 1(c) shows the FC dependence on the thickness of the Co/Pt multilayers without Ru spacer layer. The 2ML sample has the smallest switching field, and the 4ML and 6ML (or $t_{Ru} = 0$-nm SAF) samples have the similar switching field. Figure 1(d) shows the normalized in-plane and out-of-plane M–H loops of the $t_{Ru} = 0.6$ nm SAF multilayer at 300 K, which indicates an easy axis of perpendicular magnetic anisotropy. Note that long/short arrows are used in the picture to denote the magnetization direction of the upper and lower multilayers, respectively. For the in-plane M–H loop, a single-step switching is observed. While, for the out-of-plane M–H loop, a multi-step switching is observed with the magnetization in the top/bottom FM layers changing from parallel to anti-parallel to parallel, as shown in Fig. 1(d) insets. Sharp spin-flips appears at the exchange coupling field with a small net-moment at low perpendicular field.[21] The net moment should attribute to the slightly different moment of the top/bottom FM layers, which might be caused by the difference in the microstructures and interfaces of the two Pt/Co layers.

In order to study the relationship between the exchange coupling field (H_{ex}) and the thickness of Ru spacer, we measured the out-of-plane M–H loops of four samples with different thickness of Ru at room temperature. Figure 2 displays the representative M–H loops of SAF structures with Ru spacer thickness vary from 0.6 nm to 1.6 nm. Typical AFC characteristics are observed for the SAF multilayers with $t_{Ru} = 0.6$, 0.9, 1.2 nm, in contrast, a typical FC character is observed for the $t_{Ru} = 1.6$-nm SAF multilayer similar to the $t_{Ru} = 0$-nm SAF multilayer. The transition from ferromagnetic to antiferromagnetic to ferromagnetic coupling has been observed with the thickness of Ru changing from 0 nm to 1.6 nm: the transition from ferromagnetic ($t_{Ru} = 0$ nm, see Fig. 1(b)) to antiferromagnertic ($t_{Ru} = 0.6$, 0.9, and 1.2 nm, see Fig. 2) to ferromagnetic ($t_{Ru} = 1.6$ nm, see Fig. 2). Although we did not prepare samples with thickness between 0 nm and 0.6 nm, we believe that samples with thickness in this range (like, $t_{Ru} = 0.2$ nm) should exhibit ferromagnetic coupling. This can be understood from the spacer thickness dependent oscillatory AFC and FC effect in SAF multilayers. For the SAF multilayers, the interlayer exchange coupling between the two ferromagnetic layers is oscillatory antiferromagnetic and ferromagnetic coupling as found in Refs. [14] and [22]. Although a similar MH loop was observed for $t_{Ru} = 1.6$ nm and 0 nm multilayers, the ferromagnetic coupling effects are different. For $t_{Ru} = 0$ nm, there is no the Ru spacing layer, and the exchange interaction between the two ferromagnetic layers is the intralayer ferromagnetic coupling. For $t_{Ru} = 1.6$ nm, with the presence of an Ru spacing layer, the exchange interaction between the two ferromagnetic layers is the interlayer ferromagnetic coupling which is attributed to an oscillatory decay behavior of the Ruderman–Kittel–Kasuya–Yosida (RKKY) interlayer coupling depending on the thickness of the spacer layer.[18,22] For the case of $t_{Ru} = 0.6$ nm, as the field decreases from positive saturation, the magnetization of the bottom FM layer firstly switches from up to down at 5.2 kOe, owing to that the bottom FM layer stack has a smaller moment than the top FM layer as shown in Fig. 1(c). For the case of $t_{Ru} = 0.9/1.2$ nm, H_{ex} is around 3.6/2.48 kOe, lower than that of $t_{Ru} = 0.6$ nm. We can find that H_{ex} decreases with increasing the thickness of Ru for SAF multilayers with antiferromagnetic coupling properties. The interlayer exchange coupling strength ($-J_{iec}$) was determined by the equation $-J_{iec} = H_{ex}M_s t_{FM}$,[19,23–25] where the M_s refer to the saturation magnetization and the t_{FM} refer to the total thickness of both top and bottom FM layers.

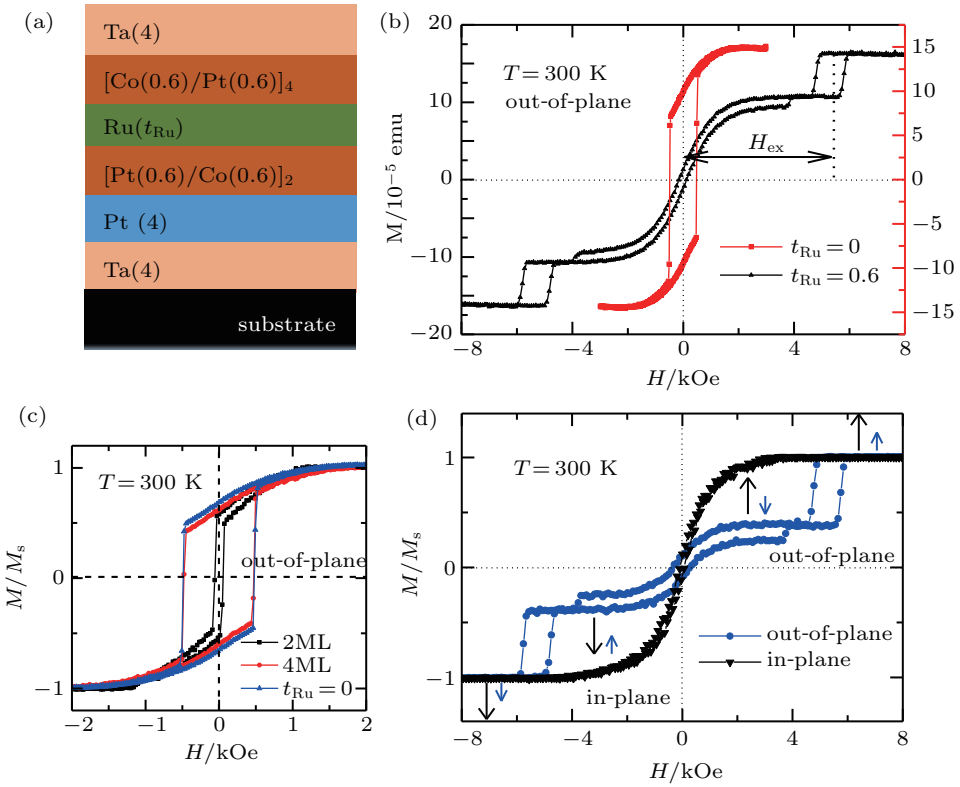


Fig. 1. (color online) (a) Schematic illustration for [Co/Pt] superlattice-based p-SAF multilayer structure. (b) The out-of-plane M–H curves of p-SAF structures with $t_{Ru} = 0.0$ nm and 0.6 nm, respectively. (c) The normalized out-of-plane M–H curves for different $[Co/Pt]_x$ superlattices ($x = 2$, 4, 6) without Ru spacer layer. (d) The normalized out-of-plane and in-plane M–H curves of the p-SAF structure with $t_{Ru} = 0.6$ nm at 300 K. The inserted long/short arrows indicate the out-of-plane magnetization in the top/bottom FM layers of the SAF multilayers, respectively.

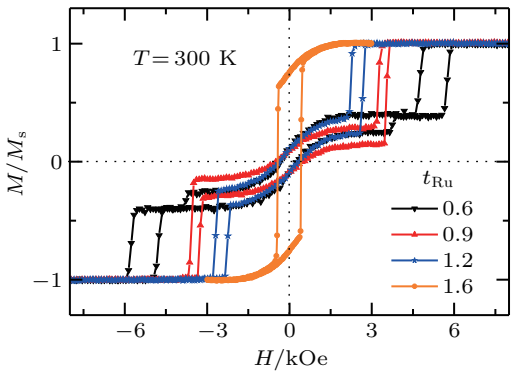


Fig. 2. (color online) The dependence of the normalized out-of-plane M–H loop on the thickness of the Ru spacing layer in the p-SAF structure at 300 K, for $t_{Ru} = 0.6$, 0.9, 1.2, 1.6 nm.

From the application point of view, the thermal stability of the interlayer exchange coupling field (H_{ex}) and strength ($-J_{iec}$) is the meaningful factor for the development of novel spintronic devices based on SAF multilayers. It has been found that the H_{ex} and $-J_{iec}$ of SAF multilayers varies significantly with temperature owing to the Fermi surface state of the spacing layer, which has been studied in previous reports.[26,27] Figure 3 shows the normalized out-of-plane M–H loops of the SAF structures with $t_{Ru} = 0.6$, 0.9 nm at different temperatures, *i.e.*, 100 K, 400 K, and 600 K. As shown in Fig. 3(a), for $t_{Ru} = 0.6$ nm, the H_{ex} is about 4.8 kOe and the $-J_{iec}$ value was evaluated to be 3.893 erg/cm^2 at 100 K. With temperature increasing, the H_{ex} increased firstly to 5.37 kOe at 400 K, and then decreases to 3.2 kOe at 600 K. However, for $t_{Ru} = 0.9$ nm, the H_{ex} decreases from 4.4 kOe to 2.1 kOe with temperature increasing from 100 K to 600 K. While, the $-J_{iec}$ decreases from 4.5 erg/cm^2 to 1.53 erg/cm^2. The degradation of H_{ex} in high annealing temperature might be caused by the diffusion of Ru atoms in the spacer layer between Co/Pt layers. We have observed a much higher value of $H_{ex} = 2.1$ kOe for $t_{Ru} = 0.9$ nm even at the high temperature of $T = 600$ K than other report[28] because of the thermally robust characteristic of Co/Pt layer.[29–31]

The temperature dependence of H_{ex} for SAF multilayers with three different Ru thicknesses are shown in Fig. 4(a). For $t_{Ru} = 0.6$ nm, a unique non-linear temperature dependence was observed with the maximum value appears around 300 K. The H_{ex} increases with increasing temperature from 100 K to 300 K. For temperature above 300 K, H_{ex} decreases with temperature increases. However, for $t_{Ru} = 0.9$, 1.2 nm, a linear temperature dependence of H_{ex} was observed. H_{ex} decreases with increasing temperature from 100 K to 600 K. The reason for the increase below 300 K is unclear but may be caused by the large coercivity of $[Pt(0.6)/Co(0.6)]_2$ layer for thinner spacer layer.[32] Figure 4(b) shows the Ru thickness dependence of H_{ex} at various temperatures. For each individual temperature of all the six measured temperatures, H_{ex} decreases

with increasing Ru thickness. For instance, at $T = 400$ K, H_{ex} decreases from 5.37 kOe to 1.9 kOe when the thickness of Ru increased from 0.6 nm to 1.2 nm.

The temperature dependence of $-J_{iec}$ for SAF multilayers with three different Ru thicknesses are shown in Fig. 4(c). The $-J_{iec}$ exhibits a similar trend as that of H_{ex} as shown in Fig. 4(a). The $-J_{iec}$ decreases with increasing temperature for $t_{Ru} = 0.9$ nm and 1.2 nm, but a nonlinear temperature dependence of $-J_{iec}$ was also observed for $t_{Ru} = 0.6$ nm. Figure 4(d) shows the Ru thickness dependence of $-J_{iec}$ at various temperatures. The $-J_{iec}$ exhibits a different trend from that of H_{ex} as shown in Fig. 4(b). For temperatures below 300 K, the $-J_{iec}$ increases with increasing the Ru thickness. However, for temperatures above 300 K, the $-J_{iec}$ decreases with increasing the Ru thickness. It is worth mentioning that H_{ex} and $-J_{iec}$ are still as high as 3.19 kOe and 1.97 erg/cm^2 for the SAF structure with $t_{Ru} = 0.6$ nm at 600 K. We also did the measurement at the temperature of 700 K, but the SAF multilayers damaged at such a high temperature.

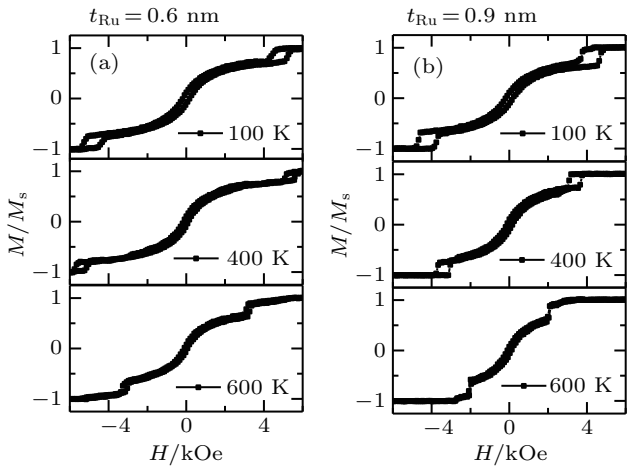


Fig. 3. Temperature dependence of the normalized out-of-plane M–H loops of SAF multilayer structures with (a) $t_{Ru} = 0.6$ nm and (b) $t_{Ru} = 0.9$ nm.

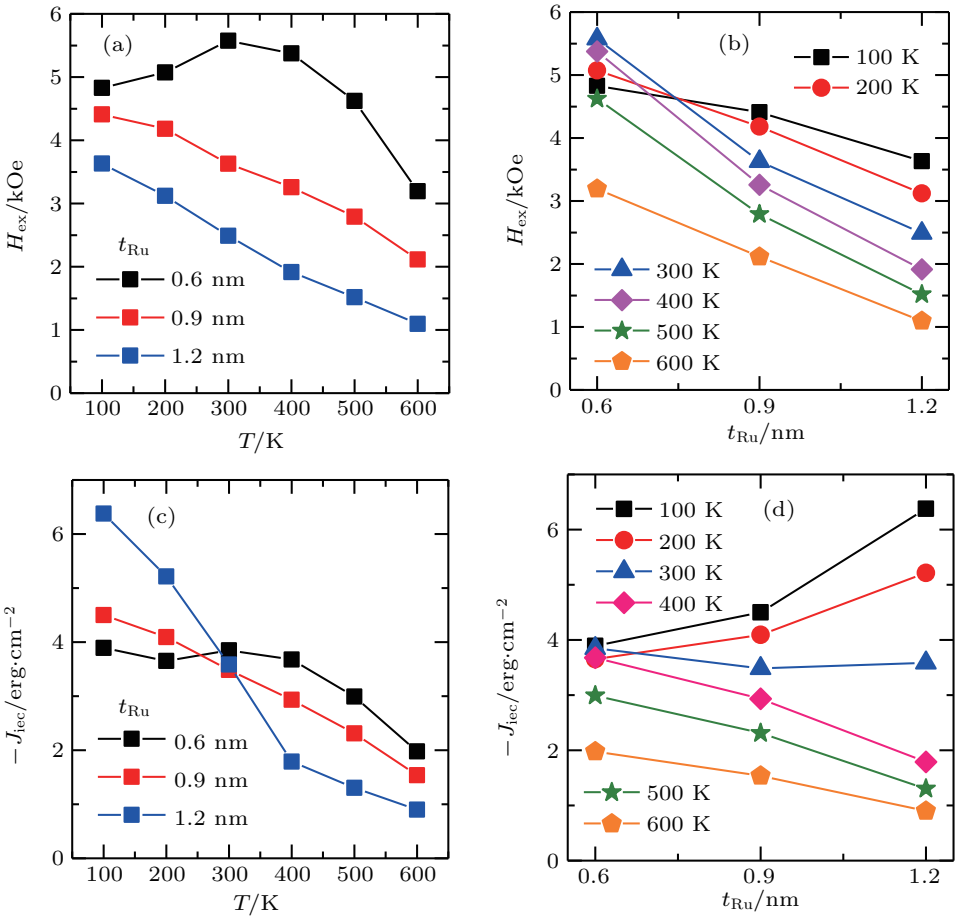


Fig. 4. (color online) Temperature dependence of the exchange coupling field (H_{ex}) (a) and the exchange coupling strength ($-J_{iec}$) (b) for SAF multilayers with three different spacing Ru layers, respectively. (b) and (d) The spacing Ru layer thickness dependence of H_{ex} (b) and $-J_{iec}$ (d) under various temperatures.

4. Summary

In this work, the temperature dependence of the interlayer exchange coupling field (H_{ex}) and coupling strength ($-J_{iec}$) in magnetron sputtered SAF multilayers with perpendicular magnetic anisotropy have been studied experimentally and theoretically. It is experimentally demonstrated that the IEC behavior of [Pt(0.6)/Co(0.6)]$_2$/Ru(t_{Ru})/[Co(0.6)/Pt(0.6)]$_4$ multilayers is very sensitive to the spacing layer Ru and the tem-

perature variation. It is worth pointing out that our SAF multilayers possess a much higher thermal stability even at the temperature of 600 K ($H_{ex} = 3.19$ kOe, $-J_{iec} = 1.97$ erg/cm^2 for $t_{Ru} = 0.6$ nm) in comparison with other reported values. The enhanced H_{ex} and J_{iec} make [Pt/Co] multilayer structure possess large perpendicular magnetic anisotropy (PMA) energy, which enable us to obtain an enlarged AF-coupled working field range, *i.e.*, a wider field range hosting AF-skyrmions. On the other hands, the large anisotropy for the development of high storage density device is valuable. Our results would provide benefit information for novel spintronic applications requiring SAF multilayers of better magnetic performance and thermal stability.

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