Temperature dependence of competition between interlayer and interfacial exchange couplings in ferromagnetic/antiferromagnetic/ferromagnetic trilayers

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(Received 17 September 2009; accepted 8 November 2009; published online 3 December 2009)

The competition between interlayer and interfacial exchange couplings is found to be temperature dependent in Co(3 nm)/AF/Fe(10 nm) trilayers with AF≡antiferromagnetic NiO, Cr_2O_3 or Cr. The temperature dependence in trilayers with AF insulating NiO or Cr_2O_3 spacer layer differs from that with AF metallic Cr. In the insulator case, the enhancement in the interlayer exchange coupling and the reduction in interfacial exchange coupling with increasing temperature results in dominating interlayer exchange coupling at high temperature. In the metallic spacer case, both the couplings decrease with increasing temperature, resulting in decoupling at high temperatures. © *2009 American Institute of Physics*. [doi:10.1063/1.3270531]

After the first observation of the antiferromagnetic (AF) coupling in Fe/Cr/Fe trilayers, the interlayer coupling has been widely investigated.[1,2] The oscillation of the coupling strength with spacer thickness has been attributed to the topology of the Fermi surface of the spacer metals.[3–5] The nonoscillatory decay of the strength of the interlayer coupling with thickness of the insulating spacer layer has been observed[6] and explained by the models.[7,8] In contrast, oscillation of the interlayer coupling was found in $[Pt/Co]_3/NiO/[Pt/Co]_3$ multilayers.[9] A noncollinear alignment of the magnetization directions of two ferromagnetic (FM) layers was found in several FM/AF/FM systems, which is due to a biquadratic coupling in the energy equation of the system.[10] Especially, a spiral spin structure of the AF layer can result in different angles between the magnetization axes of the two FM layers in FM/AF/FM trilayers.[11] A 90° interlayer coupling was observed in FM/AF/FM trilayers with NiO or Mn as spacer layer.[12–14] Among these systems,[9–14] the identical FM layers leads to a difficulty to separate the contributions of the two FM layers to the magnetization curves.[12,14] On the other hand, a spiral spin structure of the AF layer and a 90° coupling between the two FM layers could be observed when the magnetic anisotropies of the two FM layers are different.[11,13] However, in FM/AF/FM systems, only few investigations have been focused on the competition between the interfacial coupling between FM and AF layers and the interlayer coupling between the two FM layers. All previous work did not notice the competition between these two couplings.[9,11] In this letter, we report the experimental observation of this competition at different temperatures *T*, in Co/AF/Fe trilayers with the AF insulators Cr_2O_3 and NiO, and the AF metal Cr.

Three samples of Si (100) (substrate)/Pt (10 nm)/Co(3 nm)/AF/Fe (10 nm)/Pt (5 nm) trilayers with AF≡NiO, Cr_2O_3, and Cr (denoted as samples 1, 2 and 3, respectively) were prepared by dc and rf magnetron sputtering at room temperature. The growth of the films was carried out in a high-vacuum chamber equipped with multisputtering guns. The base pressure of the chamber was better than 2×10^{-7} Torr and Ar gas was kept at a pressure of 4×10^{-3} Torr during sputtering. Commercial Pt, Co, Cr_2O_3, NiO, Cr, and Fe targets with 99.99% purity were used. The crystal structure was investigated by means of x-ray diffraction with Cu K_α radiation. The magnetic properties at different temperature were measured using a superconducting quantum interference device.

The hysteresis loops measured at different temperatures after zero-field cooling (ZFC) are shown in Fig. 1 for sample 1. All loops are normalized to their saturation magnetization (M_S), and the magnetization corresponding to a step in a loop is called M_P. We define the parameter $L=M_P/M_S$. Clearly, the *M-H* curves exhibit a small step at 50 K and a linear increase in the magnetization at 100 K, respectively. A clear step is seen at 150 K but disappears for $T\geq310$ K. The different values for *L* at different temperatures are due to the effect of the interfacial and interlayer couplings in trilayers. The tendency observed above in the trilayers with NiO is similar to that with Cr_2O_3 (shown later).

For comparison, the hysteresis loops recorded at different temperatures after ZFC are presented in Fig. 2 for sample

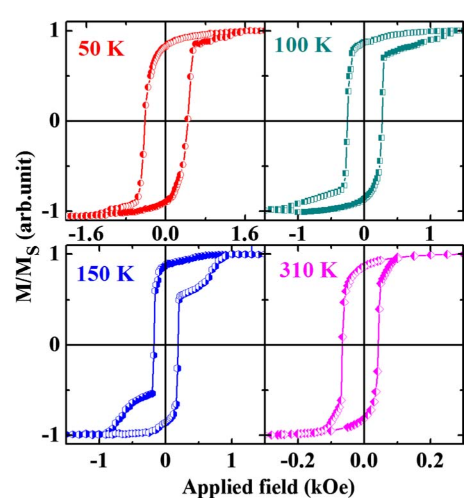

FIG. 1. (Color online) Hysteresis loops at 50, 100, 150, and 310 K of Co(3 nm)/NiO(6 nm)/Fe(10 nm) after ZFC.

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0003-6951/2009/95(22)/222505/3/$25.00

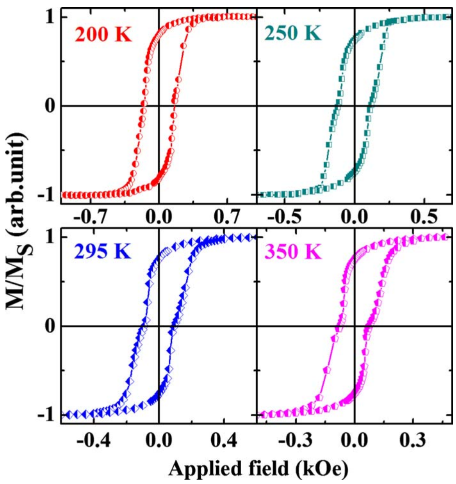

FIG. 2. (Color online) Hysteresis loops at 200, 250, 295, and 350 K of Co(3 nm)/Cr(6 nm)/Fe(10 nm) after ZFC.

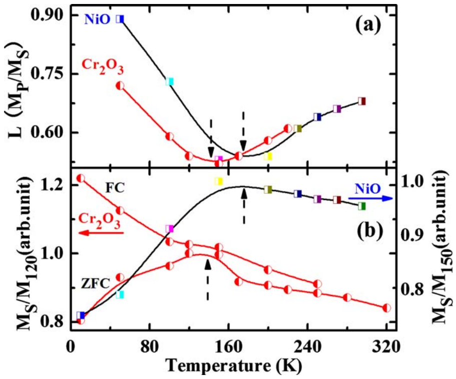

FIG. 4. (Color online) (a) Temperature dependence of L of Co(3 nm)/NiO(6 nm)/Fe(10 nm) and Co(3 nm)/Cr_2O_3(6 nm)/Fe(10 nm) after ZFC. (b) Temperature dependence of M_S/M_{120} for Co(3 nm)/Cr_2O_3(6 nm)/Fe(10 nm) after ZFC and FC, and M_S/M_{150} for Co(3 nm)/NiO(6 nm)/Fe(10 nm) after ZFC.

3. The FM layers are quite well coupled at $T=200$ K, whereas the kink observed at $T \geq 250$ K is indicative of decoupling between two FM layers. The phenomena mentioned above suggest that the variation in the couplings with temperature is quite different for the trilayers with AF materials of the two different types, i.e., insulator and metal. Furthermore, we have confirmed our results by a more systematic study on Co(3 nm)/Cr_2O_3(x)/Fe(10 nm) trilayers with $x=3$ nm, 6, 15, and 25 nm, which will be soon submitted elsewhere. However, although the experimental evidence for different behavior of insulating and metallic layers is very strong but experimental evidence is not a hard proof and it cannot totally be excluded that it is accidental.

In order to study the influence of the FC process on the magnetic properties, the hysteresis loops at 10 K of sample 2 after ZFC (black filled circle) and FC (red filled square) in an applied field of 2 kOe are presented in Fig. 3(a). For comparison, the hysteresis loop at 10 K after FC in an applied field of −2 kOe is shown in Fig. 3(b). Figures 3(c) and 3(d) show the major/minor hysteresis loops of sample 2 at 170 K and sample 1 at 200 K, respectively, both after ZFC. It is seen in Fig. 3(a) that the two FM layers switch at the same field in the ZFC loop but that a step is observed in the third quadrant of the FC loop. The value of saturation field H_S of the FC loop about −7 kOe is much larger than that of the ZFC loop, while no step is found in the first quadrant. Both hysteresis loops in Figs. 3(a) and 3(b) indicate the FM interfacial coupling between Fe and Cr_2O_3, and Co and Cr_2O_3 after FC.[15] In Figs. 3(c) and 3(d), the values of minor-loop shift $H_E<0$ indicate FM interlayer coupling in the trilayers at this temperature.[9]

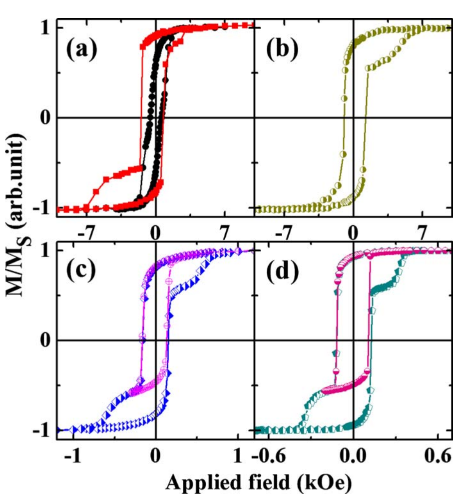

FIG. 3. (Color online) (a) Hysteresis loops at 10 K of Co(3 nm)/Cr_2O_3(6 nm)/Fe(10 nm) after ZFC (black filled circles) and FC (red filled squares) in an applied field of 2 kOe. (b) Hysteresis loop at 10 K of Co(3 nm)/Cr_2O_3(6 nm)/Fe(10 nm) after FC in an applied field of −2 kOe. (c) Major and minor hysteresis loops at 170 K of Co(3 nm)/Cr_2O_3(6 nm)/Fe(10 nm) after ZFC. (d) Major and minor hysteresis loops at 200 K of Co(3 nm)/NiO(6 nm)/Fe(10 nm) after ZFC.

The temperature dependence of L of samples 1 and 2 after ZFC (the values of L after FC are nearly constant, and are not presented here), and M_S of sample 2 after ZFC and FC and sample 1 after ZFC are shown in Fig. 4. It is found that for sample 2, the values of M_S obtained after FC decrease with increasing temperature, and are larger than those after ZFC. The same trend is seen for sample 1 (not shown here). The maxima of M_S after ZFC are found at around 120 K for the trilayers with Cr_2O_3, and at around 150 K for the trilayers with NiO. Thus, the values of M_S at different temperatures after ZFC have been normalized to M_S at 120 and 150 K for the two samples, respectively [shown in Fig. 4(b)]. It is seen in Fig. 4 that the maximum of M_S after ZFC corresponds to the minimum of L (as marked by the dashed arrows), and that the peak temperature of the film with NiO is larger than that of the film with Cr_2O_3. Furthermore, the disappearance of the step is seen at about 220 and 310 K for the film with Cr_2O_3 and NiO, respectively [see Fig. 4(a)].

The total free energy E of the Co/AF/Fe trilayers contains four contributions: (1) the interfacial coupling at two interfaces of AF layer, (2) the interlayer coupling between two FM layers, (3) the magnetocrystalline anisotropy of each layer, and (4) the Zeeman energy of each layer. The Zeeman energy of each layer and magnetocrystalline anisotropy of FM layers have little temperature dependence.[16,17] Thus, the main contributions to the energy with temperature come from (1) and (2). $J(T)=J(0)[(T/T_0)/\sinh(T/T_0)]$,[7,18] where $J(0)$ is the interlayer coupling strength at $T=0$ K. $T_0=\hbar v_F/2\pi k_B d$ is the characteristic temperature and d is the thickness of the spacer, over which the interlayer coupling strength monotonously changes with temperature. Malozemoff[19] has proposed that $H_{EX} \propto \sqrt{A_{AF}K_{AF}(0)}(1$

$-T/T_B$), where A_{AF}, K_{AF}, and T_B are the exchange constant, magnetocrystalline anisotropy, and blocking temperature, respectively, of the AF material. It is found that the interlayer coupling increases and the interfacial coupling decreases for an AF insulator but that both quantities decrease for an AF metal with increasing temperature.

At low temperatures, the interfacial coupling dominates the couplings after a ZFC process. When the interfacial coupling is strong enough, the reversal of Fe and Co layers will simultaneously occur [Fig. 3(a)]. With increasing temperature, the reduction in the interfacial coupling leads to the appearance of a step in the hysteresis loop with reduction in L. The enhancement in the interlayer coupling results in the gradual disappearance of the step and the FM coupling at $T \geq 220$ K for the film with Cr_2O_3 and at $T \geq 310$ K for the film with NiO. For the FC case, strong FM coupling in FM/AF will exist at low temperature [see the negative exchange bias field in Fig. 3(a)]. For the increasing field branch, the moments of the FM layers will reverse at a smaller field in order to reduce the interfacial energy (FM coupling in interface is in a low energy state). The stronger the exchange coupling in Co/Cr_2O_3 is, the more instable is the Co/Cr_2O_3 interface, which would lead to moment reversal of the Co and Fe layers at the same field in Fig. 3(a).[15,20] In addition, a spin-flop-like phenomenon is noticed in the film with NiO at 100 K (and with Cr_2O_3 at 50 K). Some AF materials, in which a spin-flop phenomenon may occur, have a relatively weak interaction between spin moments.[21,22] In our system, three parts can thus be assumed as an AF-like material when moments of the FM layers are antiparallel. Since the relatively weak strength of the interlayer coupling between FM layers as compared with the FM anisotropy, a magnetic field applied along the direction parallel to the easy magnetization direction of FM layers may result in a spin-flop-like phenomenon (Fig. 1). For AF insulating spacer, the interlayer coupling increases and the interfacial coupling decreases with increasing temperature. Therefore, there would be a balance between these two couplings at a critical temperature (T_{cr}). At $T < T_{cr}$, the large L and small M_S are caused by the strong interfacial coupling between AF and FM, which are also due to no orientation of magnetic moments at AF/FM interface after ZFC. The interlayer coupling becomes dominant at $T > T_{cr}$, the FM interlayer coupling will result in the increase in L, and the decrease in M_S can be due to the thermal effect. As a result, the temperature of minima of L and maxima of M_S after ZFC corresponds to this critical temperature in Figs. 4(a) and 4(b).

In summary, the interfacial coupling of Co/AF/Fe trilayers dominates the couplings at low temperatures, while the increase in interlayer coupling and the decrease in interfacial coupling lead to the domination of the former for insulating spacer layer with increasing temperature. Decoupling of FM layers is found in Co/Cr/Fe at high temperatures.

This work has been supported by the National Nature Science Foundation of China under projects 50831006 and 50971123 and National Basic Research Program (Grant No. 2010CB934603) of China, Ministry of Science and Technology of China.

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