High magnetocaloric efficiency of a NiFe/NiCu/CoFe/MnIr multilayer in a small magnetic field

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The isothermal magnetic entropy changes ΔS (i.e., the magnetocaloric potential) are studied in Ni₈₀Fe₂₀/Ni₆₇Cu₃₃/Co₉₀Fe₁₀/Mn₈₀Ir₂₀ stacks at temperatures near the Curie point of the Ni₆₇Cu₃₃ spacer by applying magnetic fields of a few milli-Tesla. Such fields were sufficient for toggling magnetic moments in the soft ferromagnetic layer (Ni₈₀Fe₂₀). It is found that this switching provides a significant enhancement of ΔS in the heterostructure system with respect to that achieved in a single Ni₆₇Cu₃₃ film under such weak magnetic fields. Our finding is believed to have the potential to be utilized in magnetocaloric devices (e.g., thin-film coolers) that would be based on ferromagnetic/paramagnetic/ferromagnetic heterostructures and would operate with moderate magnetic fields.

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I. INTRODUCTION

In materials that exhibit a strong magnetocaloric effect (MCE), high magnetocaloric efficiency can be achieved in the vicinity of phase magnetic transitions, e.g., near the Curie point $(T_{\rm C})$ [1,2]. The MCE, observed currently in advanced magnetocaloric materials, is believed to be sufficient for cooling down (or heating up) the material by applying a magnetic field up to several Tesla over several tens of thermodynamic cycles [3–6]. However, such strong fields can only be produced with bulky magnets, which are not desirable to be employed in magnetic refrigeration [7]. Therefore, there have been attempts to find MCE materials in both their bulk and structured forms [7-12] that could be used for magnetic cooling under moderate fields. We propose to enhance the MCE by surrounding a magnetocaloric material by higher- $T_{\rm C}$ (strong) ferromagnets (FM's), [13,14] so that their reconfiguration in a weak enough field could affect magnetic moments in a paramagnetic (PM) spacer due to the effect of proximity [15]. It has been shown [14,16–19] that strong FM's surrounding a PM or a weakly FM spacer are exchange-coupled through the spacer, at least up to its thickness of $\sim 20 \text{ nm}$ [14].

Here we report on our measurements of the magnetic entropy changes ΔS (magnetocaloric potential) in Ni₈₀Fe₂₀/Ni₆₇Cu₃₃/Co₉₀Fe₁₀/Mn₈₀Ir₂₀ stacks at temperatures (*T*) close to $T_{\rm C}$ of the PM = Ni₆₇Cu₃₃ spacer between the strong FM layers, i.e., FM₁ = Ni₈₀Fe₂₀ and FM₂ = Co₉₀Fe₁₀. Such a heterostructure system exhibits reconfigurations of the mutual orientation of magnetizations M₁ and M₂ in the FM₁ and FM₂ layers under an applied field of $\mu_0 H_{\rm sw} \sim 2$ mT due to their exchange decoupling across the spacer above $T_{\rm C}$ [16–19]. Our evaluations [13] of the MCE in FM/PM/FM structures anticipate a very high value of $dT/d(\mu_0 H) \approx 20$ K/T in bias fields of only a few milli-Tesla. As proposed in Refs. [13,14], this originates from demagnetization (magnetization) of the PM spacer when the mutual orientation of M₁ and M₂ alters from parallel (antiparallel) to antiparallel (parallel) alignment.

Such a reconfiguration in a FM₁/PM/FM₂/AF stack, where AF = Mn₈₀Ir₂₀ is the antiferromagnetic layer, is schematically shown in Fig. 1. The role of the AF layer is to render the FM₂ layer magnetically hard, which would contrast with the magnetically soft FM₁ layer. The dependence of ΔS on the mutual orientation of the magnetizations is associated with the giant MCE by analogy with the effect of giant magnetoresistance in spin valves [20,21]. It is also important that T_C of the spacer can be tunable by varying the spacer composition. For example, a diluted Ni_xCu_{1-x} alloy, whose T_C depends almost linearly on Ni concentration, is a good candidate for the spacer material [16–19].

II. SAMPLES PREPARATION AND CHARACTERIZATION

FM₁/PM/FM₂/AF samples were fabricated with highvacuum magnetron sputtering (AJA 2200 multichamber system) onto fused polished quartz substrates at a basic pressure of 10^{-5} Pa. To prevent them from oxidation, the samples were covered by a layer of TaO. The layers of Ni₈₀Fe₂₀ (FM₁), Co₉₀Fe₁₀ (FM₂), and Mn₈₀Ir₂₀ (AF) were sputtered from single targets, while PM films of Ni_xCu_{1-x} solid solutions were obtained by simultaneous sputtering of Ni and Cu targets, and their composition was determined by x-ray microanalysis using a dispersion spectrometer (INCA Energy Oxford Instruments). The exchange pinning between the FM_2 and AF layers was achieved during the film growth with the assistance of an in-plane magnetic field of $\mu_0 H \approx 200 \,\mathrm{mT}$. Sample magnetization as a function of H was measured with a Lake Shore 7400 Series vibrating sample magnetometer and a MPMS-XL5 Quantum Design SQUID magnetometer in a 5-400 K temperature range and external magnetic fields up to $\mu_0 H = 0.5$ T applied in the film plane. The sample area used in these studies was $A = 0.2 \text{ cm}^2$. The individual magnetizations of the strong FM layers, M_1 and M_2 , were determined from the Kittel equation by measuring the ferromagnetic resonance

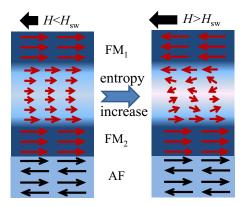


FIG. 1. Schematic of magnetic configurations in the $FM_1/PM/FM_2/AF$ heterostructure system, where AF is the antiferromagnet, whose role is to pin magnetic moments in the ferromagnet FM_2 , while the FM_1 is magnetically soft. As magnetizations in the FM₁ and FM₂ layers are oriented in the same direction at $H < H_{sw}$, where H_{sw} is the switching field for magnetic moments in the soft layer, the PM spacer is magnetized (low magnetic entropy) by exchange coupling to the FM₁ and FM₂ across the interfaces. However, the PM spacer becomes demagnetized (high magnetic entropy) after switching of magnetization in the FM_1 layer at $H = H_{sw}$. In this thought experiment, the temperature is close to $T_{\rm C}$ of the PM spacer. In the geometry of antiparallel alignment $(H > H_{sw})$ of magnetic moments in the FM₁ and FM₂ layers, according to Eq. (A5), the magnetization becomes zero at the spacer center. This can be explained by thermal excitations of magnetic moments inside the spacer under such a configuration of the magnetizations. Under the parallel configuration ($H < H_{sw}$), magnetic moments inside the spacer are not thermally excited. Therefore, magnetization switching at $H = H_{sw}$ is expected to provide an enhanced magnetocaloric potential ΔS [13,14] by analogy with the effect of giant magnetoresistance [20,21].

[14]. The composition of the Ni_xCu_{1-x} solid solution was x = 67 at. %, while the thickness of the Ni₆₇Cu₃₃ spacer (h_{NiCu}) varied from 7 to 21 nm. The thickness of the soft Ni₈₀Fe₂₀ layer (FM₁) was $h_{\text{NiFe}} = (10 \pm 1)$ nm, and these

quantities for the rest of the layers were as follows: 3.0 nm of $Co_{90}Fe_{10}$, 25 nm of $Mn_{80}Ir_{20}$, and 15 nm of TaO. The layer thicknesses were evaluated with small-angle x-ray diffraction data (reflectometry Bruker D8 Discover).

III. RESULTS AND DISCUSSION

Figure 2(a) shows the magnetization (*M*) isotherms, measured as the total magnetic moment $\mu \equiv MV$ of a TaO/Mn₈₀Ir₂₀/Co₉₀Fe₁₀/(10 nm)Ni₆₇Cu₃₃/Ni₈₀Fe₂₀/substrate sample as a function of *H*, at temperatures of T = 300, 250, and 5 K. In the observed *M*(*H*) dependences, we distinguish two steps that correspond to the switching of the soft (FM₁) and pinned (FM₂) layers. Importantly, the observed change in *MV* at the first (lower-*H*) step, which reflects switching of magnetic moments in both the FM₁ layer and Ni₆₇Cu₃₃ spacer (PM), is $A(2M_1h_{NiFe} + mh_{NiCu}) \approx 3.2 \times 10^{-7}$ A m², where *m* is the spacer magnetization. Retrieving the magnetization of the soft Ni₈₀Fe₂₀ layer $M_1 = 730$ kA/m from the ferromagnetic resonance data [14], we find that

$$2M_1 h_{\rm NiFe} >> m h_{\rm NiCu},\tag{1}$$

which is compatible with the expected smallness of magnetization in the PM spacer by comparison to that in the FM surrounding. The observed delay in the switching of magnetization in the FM₂ layer results from the exchange coupling of magnetic moments across the FM₂/AF interface. We also see that the field H_{sw} at which magnetization (**M**₁) in the FM₁ layer is switchable increases upon lowering *T*. Therefore, reversal of **M**₁ becomes undistinguishable from that of **M**₂ of the FM₂ layer at low enough *T* < 100 K.

In Fig. 2(b) we show H_{sw} versus *T* near $T_C \approx 240$ K of Ni₆₇Cu₃₃ [22] for the samples with 10- and 21-nm-thick Ni₆₇Cu₃₃ spacers. The fact that H_{sw} depends on *T* indicates that exchange through the spacer is sensitive to the temperature. Indeed, reversal of M₁ occurs at $H_{sw} = JM_2$ [23], where JM_2 is the effective magnetic field acting on the FM₁ layer from the FM₂ layer pinned by the AF, and *J* is the interlayer exchange constant, which depends on *T* [17]. Also, the J(T) dependence—with its derivation in the explicit form—is

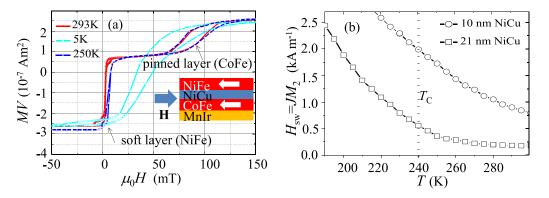


FIG. 2. (a) Isothermal magnetization of a TaO/Mn₈₀Ir₂₀/Co₉₀Fe₁₀/(10 nm)Ni₆₇Cu₃₃/Ni₈₀Fe₂₀/substrate sample at temperatures of T = 300, 250, and 5 K. In the response to an applied magnetic field ($\mu_0 H$), the magnetization reversal occurs in two steps that correspond to magnetization switching in the soft (FM₁ = Ni₈₀Fe₂₀) and pinned (FM₂ = Co₉₀Fe₁₀) layers. Reversal of magnetization in the FM₂ layer is delayed due to exchange coupling of magnetic moments in this layer with those in the antiferromagnetic layer (AF = Mn₈₀Ir₂₀) across the FM₂/AF interface. The inset illustrates the mutual configuration of an applied field **H** and magnetizations in the FM₁ and FM₂ layers before switching of magnetization in the soft FM₁ layer, which occurs at $H = H_{sw}$. (b) The switching field $H_{sw} = J(T)M_2$ [23] measured as a function of temperature T for 10-and 21-nm-thick Ni₆₇Cu₃₃ spacers.

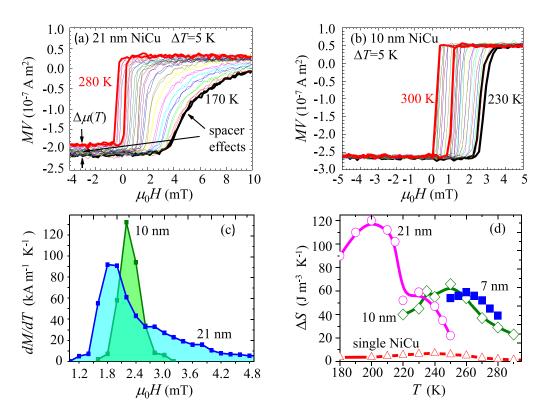


FIG. 3. (a),(b) Reversal of magnetization in the soft FM₁ layer under an applied magnetic field *H* at different *T*. The *M*(*H*) curves were taken for the samples with 21 nm (a) and 10 nm (b) Ni₆₇Cu₃₃ spacers in temperature ranges of 280–170 and 300–230 K with steps of 5 K. The spacer effects on *M*(*H*) are clearly pronounced in the sample with the thicker spacer. (c) $\partial M/\partial T$ -vs-*H* curves for the samples with 10 and 21 nm spacers at *T* = 250 and 210 K, respectively, at which ΔS are maximal. (d) $\Delta S(T)$ curves evaluated by integration according to Eq. (2) for the samples with spacers of different thicknesses – 7, 10, and 21 nm. Obtained values of ΔS for the heterostructure samples are compared to that quantity in a single Ni₆₇Cu₃₃ film, which is retrievable by extrapolation into the low-field region ($\mu_0 H \sim 2.0 \text{ mT}$) of the experimental $\Delta S(T)$ dependences measured at high $\mu_0 H \ge 1$ T and presented in Ref. [22].

discussed in the Appendix. We do not mark a large difference between H_{sw} as functions of *T* for samples with different thicknesses of the spacer.

Figures 3(a) and 3(b) show in greater detail how the soft ferromagnet (FM₁) switches in the samples with 21-nm-thick (a) and 10-nm-thick (b) spacers. These data were collected in temperature ranges of 280-170 and 300-230 K with steps of $\Delta T = 5$ K. A magnetic field **H** was applied along the direction antiparallel to that of M_1 and M_2 in their initial configuration [inset in Fig. 2(a)]. We have found that the magnetic response in both samples depends strongly on T under H in a few mT. Strikingly, in the sample with a thicker spacer, there is a contribution of the spacer to M(H), that is, the increase in saturation magnetization with lowering T, which is indicated in Fig. 3(a) by $\Delta \mu$ that depends on T. Moreover, a much more gradual switching of the FM1 in this sample, which depends on T in a relatively broad field range (up to $\mu_0 H \sim 5$ mT), can also be attributed to the spacer effect. Switching of the FM₁ in the sample with a thinner spacer occurs in a much sharper manner [Fig. 3(b)]. The spacer contribution to M(H,T) is expected to provide the magnetocaloric potential, which can be accounted for in accordance with the Maxwell relation

$$\Delta S = \mu_0 \int_{H_{\rm sw} - \varepsilon_1}^{H_{\rm sw} + \varepsilon_2} \left(\frac{\partial M}{\partial T}\right)_H dH.$$
 (2)

The $\partial M/\partial T - H$ data, required for accounting for ΔS , were obtained by plotting the M(T) and their numerical differentiation within a field range of $[H_{sw} - \varepsilon_1; H_{sw} + \varepsilon_2]$ inside of which $(\partial M/\partial T)_H \neq 0$. The $\partial M/\partial T$ -versus-H curves are shown in Fig. 3(c) for the samples with 10- and 21-nm-thick spacers at temperatures T = 250 and 210 K, which provide the largest values of ΔS in accordance with Eq. (2). It is interesting that the $\partial M/\partial T$ -versus-*H* curves are asymmetric functions with respect to H_{sw} , at which $\partial M/\partial T$ are maximal. It is seen that this asymmetry is much stronger in the sample with a thicker spacer where the spacer effects on the switching of the FM₁ are more pronounced. Because of exchange coupling at the FM₁/PM interface, thermally unstable magnetic moments inside the PM spacer intervene in the switching of the FM₁, which becomes more gradual upon thickening the spacer [Fig. 3(a)]. Therefore, the spacer contribution to ΔS is distinguishable: The more pronounced the asymmetry is in the dependence of $\partial M/\partial T$ on *H*, the higher is ΔS .

Figure 3(d) shows $\Delta S(T)$ dependences for the samples with 7-, 10-, and 21-nm-thick spacers. The magnetocaloric potential retrieved in the heterostructure samples can be compared to that quantity in a single Ni₆₇Cu₃₃ film exposed by a magnetic field of $\mu_0 H \sim 2$ mT. The values of ΔS in such weak fields for Ni₆₇Cu₃₃ were found by extrapolation into the low-field region of the $\Delta S(H) \propto H^n$ dependences [24] with $n \approx 0.8$ at *T* close to $T_C \approx 240$ K of Ni₆₇Cu₃₃, which have been reported

in Ref. [22]. Although the maximal peak values of $\partial M/\partial T$ are obtained to be highest for the sample with a thinner (10 nm) spacer [Fig. 3(c)], we mark that $\Delta S \approx 120 \text{ J/m}^3$ K is higher in the sample with a relatively thick (21 nm) spacer [Fig. 3(d)]. Strikingly, the maximal ΔS obtained in the heterostructure samples is up to ~20 times larger than that in a single Ni₆₇Cu₃₃ film. Such a change in the magnetic entropy is expected to provide the adiabatic cooling of the refrigerant $\Delta T_{ad} = -(T/C_V)\Delta S \approx -0.006 \text{ K}$, taking into account that the heat capacity increases near T_C , so that $C_V \approx 5.0 \text{ J/cm}^3\text{K}$ [26]. Note here that the magnetocaloric efficiency in the system under study is $\partial T/\partial(\mu_0 H) = -(T/C_H)(\partial M/\partial T) \sim$ -5 K/T, which is comparable to this quantity in advanced magnetocaloric materials [1,2].

It is also interesting that the magnetocaloric potential for the heterostructure system we study can also be evaluated as

$$\Delta S = 2\mu_0 (dJ/dT) M_1 M_2, \tag{3}$$

which is obtainable by substituting $M = M_1 \text{sgn}(H_{\text{eff}})$ [23] into Eq. (2). Having measured $H_{\text{sw}}(T)$ [Fig. 2(b)], we find that the highest $dJ/dT = (dH_{\text{sw}}/dT)/M_2 = 4.0 \times 10^{-5} \text{ K}^{-1}$ [25] is achievable for the sample with a thick (21 nm) spacer at $T \approx 210$ K [Fig. 2(b)], where the ΔS is largest. We see that Eq. (3) provides a simple relationship between ΔS and intrinsic properties of the spacer.

The observed enhancement of ΔS reflects the occurrence of stronger disorder in magnetic moments inside a thick enough spacer under the antiparallel configuration of M_1 and M_2 . To qualitatively explain this central feature of our work, we schematically show (Fig. 4) the isothermal magnetization of

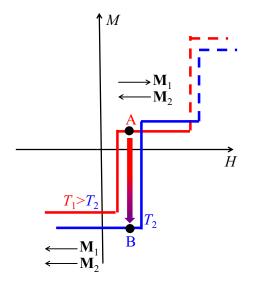


FIG. 4. Schematic of the M(H) dependence for an FM₁/PM/FM₂/AF heterostructure system at two temperatures, T_1 and T_2 . The transition of the system from the state A (antiparallel orientation of magnetizations \mathbf{M}_1 and \mathbf{M}_2) to the state B (parallel orientation of the magnetizations \mathbf{M}_1 and \mathbf{M}_2) is triggered by lowering the temperature from T_1 to T_2 . The switching between the A and B states provides the redistribution of magnetization inside the spacer and thus the change in magnetic entropy.

the $FM_1/PM/FM_2/AF$ system as a function of H for two temperatures T_1 and T_2 , so that $T_1 > T_2$ and thus $J(T_1) < J(T_2)$. If, for instance, the system stays in the state A at T_1 , lowering the temperature down to T_2 provides a transition of the system to the state B, with a strongly different magnetization, as indicated by the arrow. This transition results from the switching of M_1 , which provides the redistribution of magnetic moments in the spacer. We emphasize here that, in the geometry of antiparallel alignment of M_1 and M_2 (state A), magnetization at the center of the PM spacer is zero; see Eq. (A5). This results from thermal excitations of magnetic moments in the PM spacer under such a configuration of M_1 and M_2 (Fig. 1). However, the parallel configuration of M_1 and M_2 (state *B*) provides a higher thermal stability of magnetic moments in the spacer. Therefore, a switching of M_1 can provide the change in magnetic entropy. A temperature range within which the transition from state A to B occurs is defined by the efficiency of the interlayer exchange as a function of T, i.e., by dJ/dT. The change of the magnetic moment under varying T occurs within a finite field range, $J(T_1)M_2 < H < J(T_2)M_2$. With increasing a bias field $H > H_{sw}$, $\partial M/\partial T$ quickly decays [Fig. 3(b)]. This imposes a constraint for the adiabatic temperature change (ΔT_{ad}) per one switching of \mathbf{M}_1 . If a bias field is small, $H < H_{sw}$, there is no change in the magnetic entropy at all.

IV. CONCLUSION

We show that, at temperatures close to $T_{\rm C}$ of the Ni₆₇Cu₃₃ spacer, Ni₈₀Fe₂₀/Ni₆₇Cu₃₃/Co₉₀Fe₁₀/Mn₈₀Ir₂₀ multilayers exhibit the magnetocaloric potential ΔS , which is much higher than that in a single $Ni_{67}Cu_{33}$ paramagnet under weak magnetic fields (a few milli-Tesla). The changes in magnetic entropy occur upon the magnetic-field-driven reconfiguration of the soft FM (Ni₈₀Fe₂₀) with respect to the pinned one ($Co_{90}Fe_{10}$). Magnetization switching in the Ni₈₀Fe₂₀ layer affects the distribution of magnetic moments in the spacer [16–19], which leads to the enhancement of ΔS [13]. In this article, we present experimental evidence that the spacer is indeed responsible for this enhancement. To theoretically treat the observed changes in ΔS when altering the spacer thickness, the calculation of ΔS presented in Ref. [13] as well as the theoretical model for the interlayer exchange presented in the Appendix should be elaborated upon further. It is also important that the experimental data obtained are interpretable in a frame of a simple phenomenological model [16-19], which uncovers the relationship [Eq. (3)] between ΔS and the spacer property, i.e., the temperature derivative of the interlayer exchange constant.

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APPENDIX: SIMPLIFIED MODEL FOR INTERLAYER COUPLING

Here we derive the equation for the interlayer exchange constant and show that it is sensitive to the temperature. To describe the interlayer coupling through the PM spacer, we write the free energy per area unit [13,14]:

$$\frac{F_s}{\mu_0} = \int_{|z| < h/2} \left[\frac{1}{2} \alpha^2 \left(\frac{\partial \mathbf{m}}{\partial z} \right)^2 + \frac{1}{2} \tau \mathbf{m}^2 + \frac{1}{4m^2} \mathbf{m}^4 \right] dz + \frac{1}{2} l (\mathbf{M}_1 - \mathbf{m})^2 \Big|_{z=+h/2} + \frac{1}{2} l (\mathbf{M}_2 - \mathbf{m})^2 \Big|_{z=-h/2},$$
(A1)

where the region -h/2 < z < h/2 is occupied by a spacer with magnetization $\mathbf{m}(z)$ and is surrounded from both sides at $z = \pm h/2$ by strong FM's with magnetizations \mathbf{M}_1 and \mathbf{M}_2 for simplicity, having equal saturation magnetizations $M_1 = M_2 \equiv M$ with the components of $(M \cos \theta/2, \pm M \sin \theta/2, 0), \theta$ is the angle between magnetizations of the strong FM's, $\tau = (T - T_C)/T_C$, $\alpha \approx (k_B T_C/am_0^2)^{1/2}$ is the effective exchange length, *a* is the interatomic distance, and m_0 is the saturation magnetization of the spacer. The first term in Eq. (A1) is the free energy of the spacer, which is written in the form of the Landau expansion. The second and third terms reflect the exchange with the strong FM's that surround the PM spacer, in which *l* is the exchange constant at the interfaces.

The equation that gives extreme(s) of the functional in Eq. (A1) reads

$$\alpha^2 \frac{\partial^2 \mathbf{m}}{\partial z^2} - \tau \mathbf{m} - \frac{\mathbf{m}^3}{m_0^2} = 0, \qquad (A2)$$

while the boundary conditions at the interfaces are as follows:

$$\alpha^2 \frac{\partial \mathbf{m}}{\partial z} = \pm l(\mathbf{M}_{1,2} - \mathbf{m})|_{z = \pm h/2}.$$
 (A3)

Finding $\mathbf{m}(z)$ by solving the system of Eqs. (A2) and (A3) and substitution of the obtained solution into Eq. (A1) should allow one to clarify how the interlayer exchange depends on different parameters, e.g., ϑ , *T*, and *h*. Within an approximation of the weak exchange at the interfaces, i.e., $l << h\tau$ and $lh << \alpha^2$, one can discard the nonlinear term in Eq. (A2), so that the solution could be found in its analytic form. Then, the magnetization components in the spacer can be written as

$$m_{x} = \frac{lM\cos(\vartheta/2)}{\alpha\sqrt{\tau}\sinh(h/2\xi) + \xi\cosh(h/2\xi)}\cosh(z/\xi),$$

- h/2 < z < h/2; (A4)
$$m_{z} = \frac{lM\sin(\vartheta/2)}{\sin(\vartheta/2)}\sinh(z/\xi)$$

$$n_y = \frac{1}{\alpha\sqrt{\tau}\cosh(h/2\xi) + \xi\sinh(h/2\xi)}\sinh(z/\xi),$$

-h/2 < z < h/2; (A5)

where $\xi = \alpha/\sqrt{\tau}$ is the correlation length in the spacer. The spacer magnetization essentially depends on h/ξ . If the spacer is thick enough $(h/\xi >>1)$, the coupling between the strong FM's through the spacer exponentially approaches zero. In the opposite case of a thin spacer $(h/\xi <<1)$ and within an approximation of the weak interfacial exchange, we get that

$$\frac{F_s}{\mu_0} = -\frac{3l^2M^2}{h\tau}\cos\vartheta.$$
 (A6)

Thus, our calculations indicate that the free energy of our system can be presented in the Heisenberg-like form, i.e., $F_s = -\mu_0 J_l \mathbf{M}_1 \mathbf{M}_2$, where $J_1 = 3l^2/h\tau$ is the interlayer exchange constant obtained in units of length. As expected, the effective interlayer coupling depends on temperature as $\propto (T - T_C)^{-1}$ at *T* close to T_C .

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