Temperature-dependent magnetization in a ferromagnetic bilayer consisting of two materials with different Curie temperatures

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A structure-dependent self-consistent mean-field model has been used to calculate the temperature-dependent magnetization in ferromagnetic bilayers that consist of two materials with different Curie temperatures. The magnetization versus temperature curves $[S(T)]$ are found to be structure sensitive among the bilayer systems with simple cubic, face-centered-cubic, and body-centered-cubic structures. The Curie temperature (T_c) enhancement of the systems due to the interface exchange coupling is discussed quantitatively as a function of bilayer structure as well as the interface coupling strength. The interface effect on the layer-dependent magnetization curves $S_i(T)$ is discussed.

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The temperature dependence of the spontaneous magnetization $S(T)$ curve in a ferromagnetic material has long been an important problem in magnetism. It is usually regarded as an intrinsic property of a given material—e.g., Fe, Co, and Ni have their own $S(T)$ curves with different Curie temperatures as well as different magnitudes of magnetization, respectively. However, a $S(T)$ curve can be modified significantly if the size of the given material is shrunk down or the dimensionality of the material is reduced. It is interesting to note that the modification and manipulation of the $S(T)$ curves by the use of ultrathin films, nanowires, and nanodots turns out to be one of the most challenging issues in current magnetism research and engineering.

It has been found that the $S(T)$ curve of a given ferromagnetic material can be manipulated not only by those mentioned above but also by some extrinsic methods. Experimentally, it was observed that the Curie temperature of Gd layers on Fe(100) was enhanced significantly from its intrinsic bulk value by the proximity effect of Fe to the Gd layers.[1,2] Similar results were also obtained for Fe films on Pd(100) (Refs. 3 and 4) and for Ni layers in the Co/Cu/Ni trilayer system with an adjustable interlayer coupling.[5] A clear physical mechanism is needed to explain all these experimental observations. Theoretically, for a bilayer or a superlattice that consists of two materials with different Curie temperatures and a ferromagnetic interface coupling, phenomenological approaches have been carried out by use of Landau-Ginzburg theory[6,7] which predicted that the magnetic susceptibility should have a single maximum (singularity) if the films are thin but have two maxima (one maximum at lower temperature, one singularity at higher temperature) if the films are thick. Meanwhile, microscopic approaches have also been developed. Camley and Li used the self-consistent local mean-field theory[8] to address the magnetic bilayers or superlattices with an antiferromagnetic interface coupling. Jensen *et al.* used the Green's function theory with a random phase approximation (RPA) to explain quantitatively the effective "Curie temperature" enhancement of Ni films in the Co/Cu/Ni trilayer system,[9] a theory that picks up part of the spin fluctuations but is still a kind of mean-field theory, which works quite well for the whole temperature range except in the vicinity of real Curie temperature.[10] Compared to the phenomenological approaches, the advantage of the microscopic approaches lies in the fact that they deal with the underlying microscopic structures in the films and the results can be in principle verified by modern element-specific techniques. For example, with the probe layer technique, Mössbauer spectroscopy is able to study the element-specific and position-selective spin arrangement.[11–13] Meanwhile, x-ray magnetic circularly dichroism (XMCD) spectroscopy using synchrotron radiation has been proven to be an even more powerful technique.[14]

In spite of the many progresses listed above, there are still some interesting issues that need to be addressed. (1) There is still lacking a microscopic model with real lattice structures of a bilayer, which consists of two materials with different Curie temperatures. For example, what is the difference of a $S(T)$ curve if the bilayer has a face-centered-cubic or a body-centered-cubic structure? For this purpose, the in-plane contribution must be included in the model Hamiltonian; otherwise, only the body-centered-cubic structure along the $\langle 100 \rangle$ directions can be treated.[8] (2) Most of the previous works focused only on the enhancement of the effective T_c of the ferromagnetic bilayers by the interface coupling, yet little attention has been paid to the change, especially the quantitative change, of the real T_c, although an increase is predicted by a theorem.[15] (3) It is not clear how far the influence of interface coupling can be extended to both sides in a ferromagnetic bilayer system, while the Mössbauer or other experimental techniques like XMCD might be able in principle to provide such information in experiments.

In this work, we develop a method that takes into account the in-plane contribution, which can be applied to study any real lattice structures of a ferromagnetic bilayer that consists of two materials with different Curie temperatures; we also describe quantitatively the enhancement of the real T_c in a magnetic bilayer, as a function of the bilayer structure as well as the interface coupling strength; we finally provide the layer-dependent $S_k(T)$ curves for each atomic layer in a bilayer system. All the results worked out here can be in principle verified by future experiments.

It should be pointed out that the magnetization versus temperature curves are the main concern of the present work, while many other interesting properties of magnetic bilayers or multilayers, such as the interface magnons,[16] spin-wave resonance,[17] the finite-size scaling of the Curie temperature,[18] and the spring magnets,[19] are not discussed here.

The system to be considered here is as following: a bilayer with film A on the top of film B, stacked along the $\langle 001 \rangle$ direction. Film A has k_1 and film B k_2 atomic layers, respectively. Define the top layer of film A as the first layer, then its bottom layer at the interface becomes the (k_1)th layer, meanwhile the first layer of film B at the interface is the (k_1+1)th layer and the bottom layer of film B is the (k_1+k_2)th layer.

Similar to that adopted by Camley and Li[8] and Jensen *et al.*[9] a three-dimensional Heisenberg Hamiltonian is used in the following to describe the magnetic bilayer with a ferromagnetic interface coupling:

$$E^{total} = -\sum_{\alpha,\beta(\alpha\neq\beta)} J^a_{\alpha\beta} S^a_\alpha \cdot S^a_\beta - \sum_{\alpha,\beta(\alpha\neq\beta)} J^b_{\alpha\beta} S^b_\alpha \cdot S^b_\beta - \left(\sum_\gamma J^{ab}_\gamma S^a_\gamma \cdot S^b_\gamma \right)_{interface}.$$

Here, the first and second terms represent the spin-spin interactions in films A and B, respectively, while the third gives the spin-spin interaction between films A and B.

Under the standard mean-field approximation, this Hamiltonian can be decoupled to the product of H_k and S_k, where $S_k=(1/N)\Sigma_{ij}S_{ijk}$ takes the kth layer as a single atomic spin whose magnitude is averaged up among the N spins and $H_k=(1/N)\Sigma_{ij}H_{ijk}$ is the averaged effective field felt by the kth layer. H_{ijk} represents the corresponding field applied to the spin at (i,j,k). N is the total spin number in each atomic layer. Thus the thermal average of the layer-dependent atomic spin at any finite temperature T can be written as

$$\langle S_k(T) \rangle = S B_s(y), \tag{1}$$

where

$$B_s(y) = \frac{2S+1}{2S} \coth \frac{2S+1}{2S} y - \frac{1}{2S} \coth \frac{y}{2S},$$

$$y = \frac{S}{kT} H_k,$$

and S is the spin angular momentum to be considered. Correspondingly the effective magnetic field is

$$H_k = z_\perp (J_{k-1}\langle S_{k-1} \rangle + J_{k+1}\langle S_{k+1} \rangle) + z_\parallel J_k \langle S_k \rangle, \tag{2}$$

where $z_\parallel$ and $z_\perp$ are the in-plane and out-of-plane nearest-neighbor coordination numbers for any specific lattices, respectively. For the three specific lattices to be treated in this work, $z_\parallel=4$ $z_\perp=1$ for simple cubic (sc), $z_\parallel=0$ $z_\perp=4$ for bcc, and $z_\parallel=4$ $z_\perp=4$ for fcc structures, respectively.

It is clear that we have now $2(k_1+k_2)$ equations from the foregoing equations (1) and (2). These $2(k_1+k_2)$ equations with (k_1+k_2) unknown spins $\langle S_k \rangle$ and (k_1+k_2) unknown magnetic field H_k can be solved self-consistently in the iterative way.

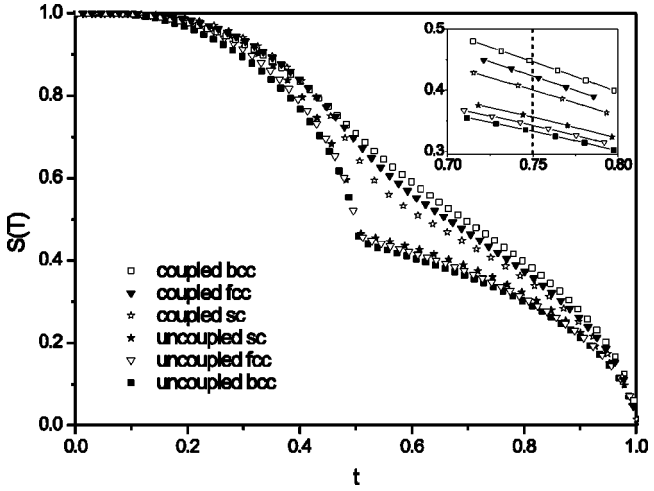

FIG. 1. Normalized $S(T)$ curves for bilayers with simple cubic, body-centered-cubic, and face-centered-cubic structures, respectively, when the interface coupling is turned on or off.

Without the loss of generality, we take $S_A=S_B=1$. It is further assumed that the exchange constants in the two free-standing films (A) and (B) are J_A and J_B, respectively, and the interface coupling constant between (A) and (B) is J_{AB}, while their relative magnitudes are set to be $J_A:J_B=1:2$, and J_{AB} is adjustable between J_A and J_B. It is clear that we are considering here two ferromagnetic films with $T_{cA}<T_{cB}$, respectively. To illustrate most clearly the interface effect to both films, 5 monolayers (ML) each for films (A) and (B) is found to be the best choice.

Figure 1 shows a set of spontaneous magnetizations as a function of temperature for the bilayers with sc, bcc, and fcc structures, respectively, when the interface coupling is turned off and on. Here J_{AB} is set to be $J_{AB}=J_B$. The absolute numerical values of J_B for the uncoupled and coupled sc, bcc, and fcc cases have been determined by normalizing the corresponding T_{cB} to 1. To avoid confusion, we use t to represent the temperature after the normalization process. It is seen that there are two independent phase transitions at $t=0.5$ and $t=1.0$, respectively, for the uncoupled cases, while there is only one real phase transition at $t=1.0$ for the coupled cases. In addition, for the uncoupled cases at any particular temperatures (see the inset) it is found that $S(T)$ of sc is at the top and bcc is at the bottom, while fcc is sandwiched in between. However, this ordering is just upside down after the coupling is turned on, where bcc sits the highest, sc the lowest, and fcc in between. It should be noted that the two sets of orderings do not correspond to the bulk coordination numbers (z_{bulk}) of the three lattices—i.e., 12 for fcc, 8 for bcc, and 6 for sc structures—as one might naively think. To understand the physics behind this, one needs to go a little deeper into this problem. For the uncoupled cases, the influences by the coordination numbers for different structures have already been automatically taken into account in the T_c normalization process. After that, the curve shapes of the three different bilayer systems reflect directly the competition between the contributions from the in-plane and the

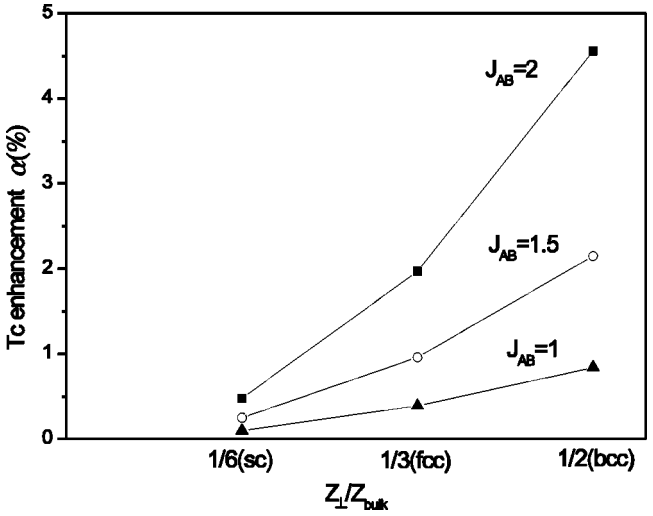

FIG. 2. T_c enhancements for bilayers with simple cubic, body centered cubic, and face centered cubic structures respectively. $\alpha=(Tc_2-Tc_1)/Tc_1$ where Tc_1 is Tc of uncoupled bilayer and Tc_2 is Tc of the coupled bilayer.

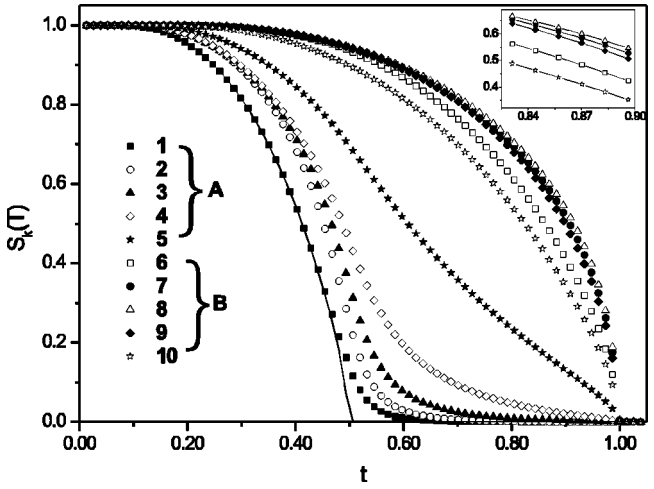

FIG. 3. Layer-dependent magnetization for a coupled bilayer with simple cubic structure. For any fixed temperatures, the layer-dependent curves of film A are ordered from the bottom as layers 1, 2, 3, 4, 5, respectively, while the curves of film B are ordered as layers 10, 6, 9, 7, 8, respectively. The solid line is the $S(T)$ curve of layer 1 in the uncoupled case.

interlayer couplings. For the (100) thin films with the nearest-neighbor exchange interaction, bcc has zero in-plane coordinates, but has eight or four interlayer coordinates depending on whether or not this layer sits in bulk or at the surface. Similarly, fcc has four in-plane coordinates but eight or four interlayer coordinates accordingly, and sc has four in-plane coordinates but two or one interlayer coordinates.

With these numbers in mind, it is not difficult to judge now that for a free-standing film with only 5 ML thick—i.e., only three layers in bulk but two layers at surfaces—the perturbation by the surfaces for the spontaneous magnetization is obviously most severe for the bcc case, but less for the fcc case and the least for the sc case. On the other hand, for the coupled cases, since the interface coupling is chosen to be the same as that in the (B) film (higher-T_c material), it is then a strong perturbation for the (A) film (lower T_c material). Therefore it is expected that the curve ordering will be reversed because the perturbation is the strongest for the bcc, less stronger for the fcc, and the weakest for the sc cases. It is thus concluded that it is the competition between the surface and interface effects that determines the detailed shape of a normalized $S(T)$ curve for any particular bilayer systems. It should be mentioned that the overall features discussed above are not changed when the interface coupling strength is shifted to that of the (A) film (lower T_c material).

Now we turn to the problem of the Curie temperature enhancement by the interface coupling. We concentrate here only on the enhancement of the Curie temperature in the real T_c. What we have done is to fix J_B and calculate T_c with interface coupling on and off. In Fig. 2, the T_c enhancement is plotted, with different interface coupling strengths, as a function of the relative interlayer coordination numbers—i.e., the ratio of coordination numbers between interlayer and bulk values—for the sc, bcc, and fcc bilayers, respectively. It is found that the degree of T_c enhancement depends on the detailed bilayer structure. The effect is the largest for the bcc structure, intermediate for fcc, and the smallest for sc. This can be interpreted as following: on the one hand, for the sc case the coordination number contributed to the enhancement of the Curie temperature increases only one per interface atom compared to the uncoupled case, while for the fcc and bcc cases the numbers are four per atom; on the other hand, these increases of coordination numbers relative to their uncoupled cases (5 for sc, 8 for fcc, and 4 for bcc structures) are quite different. It is these two factors that lead to the difference of the Curie temperature enhancement. Interestingly, if only two layers are concerned ($k_1=k_2=1$), an analytical formula of the critical temperature can be obtained explicitly from Eqs. (1) and (2) by replacing the Brillion function with the leading term of its Taylor series expansion:

$$kT_c=\frac{S(S+1)}{6}\{z_{\parallel}(J_A+J_B)+\sqrt{z_{\parallel}^2(J_A-J_B)^2+4z_{\parallel}^2J_{AB}^2}\}.$$

It is seen that any nonzero J_{AB} results in an enhanced T_c, while the degree of enhancement does depend on the structure as well as the interface coupling.

Finally we try to study how the interface coupling affects the magnetization away from the interface. We take the bilayer with a simple cubic structure as an example in the following but the results are generally valid for any other structures. Figure 3 shows the layer-dependent magnetization of the bilayer when the interface exchange coupling is turned on between films A and B. It is found that the phase transition that previously happened at $t=0.5$ does not exist anymore: meanwhile, there is still a clear phase transition at $t=1$. However, the difference compared to the uncoupled case is that there are now ten distinguished curves for the whole bilayer system, since the first and fifth layers the second and fourth layers of each film are not anymore identical after the interface coupling is turned on. For any fixed temperatures, the layer-dependent curves of film A are ordered as layers 1, 2, 3, 4, 5, respectively, starting from the bottom.

The curve from the interface layer (fifth layer) is most strongly affected. Compared to the uncoupled case, all the curves of film *A* show clear tails as the magnetization is decreased to zero, even for the farthest layer from the interface (first layer) where the tail is still obvious. As a reference to see the tail, we also put in this figure the curve of the first layer in the uncoupled case. It should be noted that these tails are very similar to that observed at the Curie point for a ferromagnetic-paramagnetic phase transition under an external magnetic field. In this sense the interface coupling acts as a kind of effective residual magnetic field for film *A*. The interface effect, however, is much weaker in film *B* as seen in Fig. 3, although the set of curves are also split to five different curves. The ordering of the curves in film *B* at any given temperature is found to be qualitatively different from that of film *A* as discussed previously. The curves of film *B* from the bottom are 10, 6, 9, 7, 8, respectively. The magnetization of the interface layer (sixth layer) is larger than the surface layer (tenth layer) since the former is coupled directly to another magnetic system. Similarly the magnetization of the seventh layer is larger than the ninth layer. The magnetization of the ninth layer is larger than the sixth layer because the former has large exchange couplings at both sides, while the latter has one side large (with the seventh layer of film *B*) and the other side small (with the fifth layer of film *A*) exchange couplings. Following the same reasoning, the magnetization of the eighth layer is the largest as expected. At last, it should be mentioned that 10 ML/10 ML and 20 ML/20 ML cases (not shown here) have also been investigated. The result shows that the interface effect on an $S(T)$ curve extends at least to the tenth layer of the lower-T_c side but is almost negligible in the second layer at the higher-T_c side.

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