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Magnetic-dipole mechanism for biquadratic interlayer coupling

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A mechanism resulting in biquadratic interlayer coupling is proposed and analyzed theoretically. This mechanism is connected with the magnetic-dipole field, created by magnetic layers with roughness. This field decays exponentially with the distance from the layer, but it shows oscillating behavior in the lateral direction. The scale of both exponential and oscillating dependencies corresponds to the scale of the interface roughness. The oscillating variation of the field makes 90° alignment of the magnetization energetically favorable in analogy to the Slonczewski mechanism. Computer simulations and estimates show that this mechanism can provide a coupling strength of the order of 0.01 erg/cm^2 for Fe films with 1 nm interlayer thickness.

I. INTRODUCTION

Since the recent discovery of a biquadratic type of interlayer coupling[1] in Fe/Cr/Fe epitaxial structures the effect was also found in other systems.[2-4] In the experiments a situation where the magnetization of two magnetic films are perpendicular to each other has been observed. This finding can be explained by assuming that the interlayer coupling energy per unit area has the phenomenological form

$$E_s = -J_1\cos\theta - J_2\cos^2\theta \ . \tag{1}$$

Here θ is the angle between the saturation magnetizations of the two films, J_1 and J_2 are the bilinear and biquadratic coupling parameters. Various mechanisms which describe this biquadratic coupling have been proposed[5-8] since its experimental observation. Slonczewski[5] related biquadratic coupling to the two-monolayer (ML)-period oscillation of the bilinear exchange. Due to this oscillation every monatomic step in a rough surface creates an area of the exchange with the opposite sign of the coupling between adjacent magnetic films. The competition between ferromagnetic and antiferromagnetic bilinear coupling together with the exchange stiffness of the magnetic layers provide the energetic preference of the magnetizations of the two magnetic films to be aligned perpendicular to each other. In this approach the strength of the biquadratic coupling J_2 is connected with that of the bilinear one, J_1. However, experimental data obtained for different systems, in particular the temperature dependencies of J_1 and J_2 cannot be explained by this mechanism. Attempts to consider an intrinsic origin of the biquadratic coupling[6-9] give very small values of the coupling strength relative to the bilinear one and predict a very fast decrease of J_2 with increasing interlayer thickness. This is not observed in the experiments. Recently a so-called "loose spin" model was proposed in which indirect exchange through unpaired spins is considered.[9]

In the present paper we propose a mechanism of biquadratic coupling caused by the magnetic-dipole field created by rough magnetic films. This field changes sign with a period corresponding to a typical scale of the film roughness in the lateral direction and favors a 90° alignment of the magnetic moments of the two magnetic layers.

II. THEORY

For a demonstration of the mechanism let us consider a two-dimensional infinite ferromagnetic layer magnetized in one plane. In the framework of a continuous approach this layer does not produce a magnetic field outside itself. However, the presence of the atomic structure of the matter and localization of magnetic spins results in a magnetic field outside the layer. Using a computer calculation, it was shown[10] that for a square lattice at a given distance z from the layer the field is spatially periodic with the lattice constant a and that each harmonic of this periodic function decays exponentially from the plane. Since the dipole interaction is of long range, a direct summation of the dipole fields is extremely time consuming, especially if one tries to take roughness into account. Therefore, it seems useful to perform an analytical transformation by expanding the field from the lattice of magnetic moments in a series of exponential terms. The square lattice of moments magnetized in plane (x,y) in the x direction produces a field, the x-projection of which can be written as follows:

$$H_d(x,y,z) = -\frac{8\pi^2\mu}{a^3}\left\{\sum_{k=1}^{\infty} k\cos\left[\frac{2\pi}{a}kx\right]\exp\left[-\frac{2\pi}{a}kz\right] + 2\sum_{k,l=1}^{\infty}\frac{k^2}{\sqrt{k^2+l^2}}\cos\left[\frac{2\pi}{a}kx\right]\cos\left[\frac{2\pi}{a}ly\right]\exp\left[-\frac{2\pi}{a}\sqrt{k^2+l^2}\,z\right]\right\} . \tag{2}$$

Here μ is the magnetic moment and a denotes the lattice constant. Derivation of this formula is given elsewhere.[11]

Using Eq. (2) one can easily take into account the roughnesses of the surface. Let us consider a layered system consisting of two magnetic films with rough interface separated by a nonmagnetic spacer. If the roughness of the second film is correlated with the roughness of the first one, one can obtain an effect which favors parallel or antiparallel alignment of the magnetizations of both layers.[12] If roughnesses of the two films are not correlated, one can take them into account separately. For simplicity let us suppose that the surface of one ferromagnetic film has an array of infinitely long growth terraces and valleys with a period L, the second film having a smooth surface. Figure 1 illustrates the situation under consideration. The bottom film consists of a set of complete layers and a few incomplete layers establishing terraces. As it is clear from Eq. (2) the dipole field of a completed layer falls off exponentially with the distance z and for $z > a$ is negligible. Therefore one has to consider the magnetic field produced by the terraces only. The field can be calculated by a summation of the dipole fields from the square lattices with lattice constant L, shifted relative to one another. Using (2) one can obtain for $z \geq a$ and $L \geq 4a$:

$$H_d(x,z) = -\frac{8\pi M\delta}{L}\sum_{m=1}^{\infty}(-1)^{m-1}\cos\left[\frac{2\pi}{L}(2m-1)x\right] \times\exp\left[\frac{2\pi}{L}(2m-1)z\right], \quad (3)$$

where $M = 2\mu/a^3$ is the magnetization of magnetic films having bcc structure. A similar result has been obtained for the magnetic-dipole field created by a stripe domain structure with perpendicular magnetization.[13] The calculated field profile of such an array of terraces at various distances z from the surface is shown in Fig. 2. As it was explained before such an alternative field results in biquadratic coupling. The interaction involved is not the interface one only, as it was considered in Ref. 5. Instead, one can introduce the surface interaction constant J_2 with dimensionality of erg/cm^2, which is now the bulk coupling strength integrated over the magnetic film thickness.

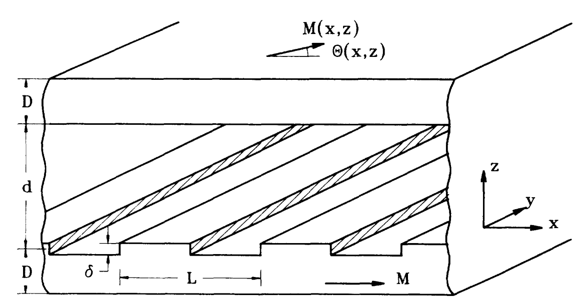

FIG. 1. Perspective section of two magnetic films separated by a spacer. The bottom film is supposed to have periodic interfacial terraces with a period L and a height δ and with its magnetization supposed to be aligned in x direction. The upper film is assumed to have a smooth interface and its magnetization forms an angle θ with the x direction. Both magnetic films have equal thicknesses D, while the thickness of the spacer layer is d.

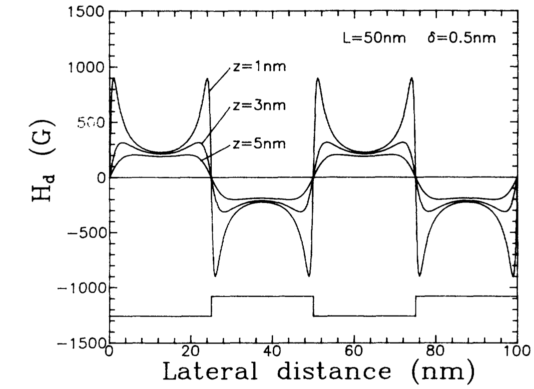

FIG. 2. Calculated profile of the magnetic-dipole field, created by terraces at various distances z from a single magnetic film for $L = 50$ nm and $\delta = 0.5$ nm. The line in the lower part of the figure indicates the position of the terraces.

The sum of energies due to intralayer ferromagnetic stiffness, cubic magnetic anisotropy and interlayer coupling caused by the magnetic-dipole field per unit area is written as

$$E_s = \frac{1}{L}\int_0^L\int_d^{d+D} dx\, dz\{A(\theta_x^2+\theta_z^2)+\tfrac{1}{4}K_1\sin^2 2\theta - MH_d(x,z)\cos\theta\}\ . \quad (4)$$

Here A is the interlayer stiffness constant, K_1 is the cubic anisotropy constant, $H_d(x,z)$, given by Eq. (3), is the calculated stray field, subscripts x and z indicate partial derivatives, and the definitions of d, D, and θ are given in Fig. 1. We did not include in (4) the energy of the stray field caused by inhomogeneous distribution of θ. Estimates show that this contribution is negligible for typical parameters characterizing experiment. Minimization of the energy with respect to θ gives us the following equation for $\theta(x,z)$:

$$2A(\theta_{xx}+\theta_{zz}) - K_1\sin 4\theta - MH_d(x,z)\sin\theta = 0 \quad (5)$$

together with the boundary conditions

$$\theta_z(x,d) = \theta_z(x,d+D) = 0\ . \quad (6)$$

Assuming that the dipole interaction results in a small change of θ relative to the average value $\bar{\theta}$, i.e.,

$$\theta(x,z) = \bar{\theta} + \delta\theta(x,z)\ ,$$

one can find an exact solution of Eqs. (5) and (6) and can express the minimum value of E_s as a function of $\bar{\theta}$:

$$E_s = \frac{1}{2\pi}\frac{M^4\delta^2 L}{\tilde{A}}\sum_{m=1}^{\infty}\frac{(-1)^{m-1}}{(2m-1)^3}\exp\left[-\frac{4\pi d}{L}(2m-1)\right] \times\left\{1-\exp\left[-\frac{4\pi D}{L}(2m-1)\right]\right\}\cos^2\bar{\theta}, \quad (7)$$

where

$$\tilde{A}=A+\frac{K_1L^2}{2\pi^2}\ .$$

Taking into account that the roughnesses of both interfaces of the two films will give contributions to J_2 independently, and comparing (7) and (1) one obtains

$$J_2=\frac{1}{\pi}\frac{M^4\delta^2L}{\tilde{A}}\sum_{m=1}^{\infty}\frac{(-1)^{m-1}}{(2m-1)^3}\exp\left[-\frac{4\pi d}{L}(2m-1)\right]\times\left\{1-\exp\left[-\frac{8\pi D}{L}(2m-1)\right]\right\}\ . \tag{8}$$

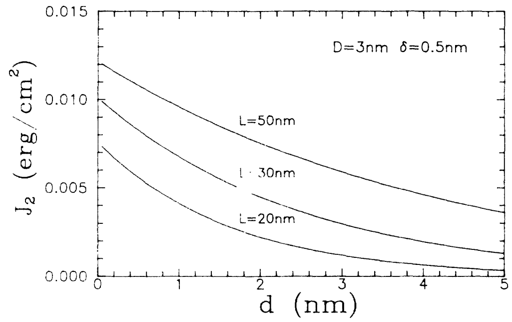

FIG. 3. Calculated value of J_2 as a function of spacer thickness d for magnetic film thickness $D=3$ nm, $\delta=0.5$ nm, and various L.

III. DISCUSSION

From Eq. (8) it becomes obvious that the strength of the coupling depends on the characterizing scale L of the interface roughness. The actual morphology of an interface in layered magnetic systems can be studied, e.g., by means of spot profile low-energy electron-diffraction analysis,[14] by direct observation of the film surface by scanning tunnel microscopy,[15] by refinement of x-ray diffraction data,[16,17] and high-resolution transmission electron microscopy combined with simulation methods.[18,19] These techniques give 5–40 nm as a typical lateral size of growth islands for molecular-beam epitaxy grown and sputtered films of 3d metals. It corresponds to $L=10$–80 nm in our notation.

The height of the roughness structure varies considerably for different samples. It may be estimated from a fast increase of the ferromagnetic coupling at the definite spacer thickness where magnetic bridges across the interlayer can appear. On the other hand, it was claimed that the absence of film roughness is closely connected with the possibility to observe a short-period oscillation of the coupling as a function of spacer thickness.[20] For samples where a short-period oscillation is damped out [e.g., Fe/Cr/Fe system, prepared at room temperature[21] or Fe/Al/Fe (Ref. 3)] both estimates show that the typical height of the roughness structure is of the order 0.5 nm.

Therefore, in case of Fe-based sandwiches one can use $L=50$ nm, $\delta=0.5$ nm in Eq. (8), as well as typical data used in experiment, $d=1$ nm, $D=3$ nm, and with known bulk values of $A=2\times10^{-6}$ erg/cm and $K_1=4.5\times10^5$ erg/cm^3. In this case we find $J_2=0.01$ erg/cm^2 and a corresponding saturation field of $H_S=18$ Oe. These values are smaller than those observed experimentally for the Fe/Cr/Fe system, but they are on the order of the value of the coupling found of Fe/Au/Fe.[3]

Figure 3 shows that the dependence of J_2 on the interlayer thickness d for different values of L. The strength of biquadratic coupling drops very slowly with d. Such a behavior agrees with experimental observation. In Ref. 1 the authors noted that in the Fe/Cr/Fe system $J_2/J_1=0.1$ for $d_{Cr}\approx1$ nm, $J_2/J_1=0.3$ for $d_{Cr}\approx2$ nm, and $J_2>J_1$ for $d_{Cr}>4$ nm. This fact indicates that with increasing spacer thickness the bilinear coupling decreases much faster than the biquadratic one. One can observe the same tendency from the experimental magnetization curves presented in Ref. 3 for the Fe/Al/Fe and Fe/Au/Fe systems. On the basis of the magnetic-dipole mechanism this fact can be explained by the different microscopic origins for the bilinear and biquadratic coupling terms in Eq. (1).

It is seen from Fig. 3 and also from Eq. (8) that J_2 increases with increasing L. It should be noted, however, that based on Eqs. (4) and (5) there exists an energy minimum only for relatively small values of L (in the case of the Fe-based system one needs $L<100$ nm). For larger values of L a stripe domain structure with head-to-head domain walls, which reflects the terraces and the valleys is energetically more preferable.

In Fig. 4 the saturation field H_S versus thickness D of the ferromagnetic films is shown. It is known that for surface interactions H_S is proportional to $1/D$. Since the magnetic-dipole coupling is caused by a bulk interaction, Fig. 4 reveals a more complicated variation of H_S. The magnetic-dipole mechanism for the biquadratic coupling also explains the fact that in many systems with different spacer materials a similar value of J_2 is observed for large values of the spacer thickness.

We should emphasize that a very important aspect of the problem is missing now in our treatment. It is known that there is interdiffusion of magnetic atoms into the spacer material.[14,15] The interdiffused atoms created paramagnetic impurities near the surfaces of the magnetic films. Due to its high magnetic susceptibility, such clusters should enhance the dipolar field inhomogeneity

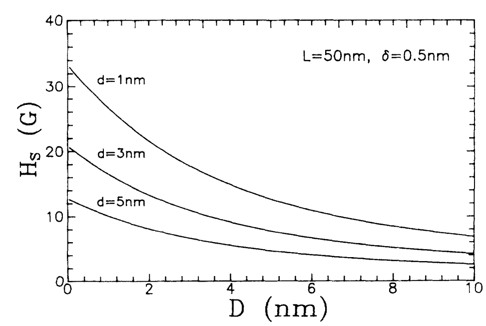

FIG. 4. Calculated value of the saturation field H_S as a function of the magnetic film thickness D for $L=50$ nm, $\delta=0.5$ nm, and for various spacer thicknesses d.

in the lateral direction. This effect will increase the value of the dipolar field and, therefore, the resulting biquadratic coupling will increase as well. Also from Curie's law, this enhancement should cause a steep temperature dependence of the strength of biquadratic coupling as is observed experimentally.[4] Examination of such a phenomenon is a subject of future investigations.

An important feature of the mechanism under discussion is that the strength of the coupling is independent of the spacer material, but it depends on the film roughness. For applications (e.g., those based on large magnetoresistance effect) it is essential to have a definite material as a spacer, which provides a way to create biquadratically coupled systems even with spacer materials for which the effect is intrinsically absent.

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