Fluctuation Mechanism for Biquadratic Exchange Coupling in Magnetic Multilayers

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We show how spatial fluctuations of chromium thickness amounting to one atomic monolayer account phenomenologically for the biquadratic magnetic coupling recently reported in epitaxial Fe/Cr/Fe trilayers. Corresponding fluctuations of the short-period oscillatory term of conventional exchange coupling induce static waves of magnetization whose energy has the observed biquadratic form. An existing calculation of conventional exchange and its recent direct measurements in highly ordered trilayers support our interpretation.

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The exchange energy which couples two or more ferromagnetic films separated by thin nonferromagnetic interlayers is currently under intense investigation. Recently attention has turned to experiments [1,2] and theory [3,4] dealing with Fe magnets separated by Cr interlayers varying between zero and many monolayers in thickness. The phenomenological coupling energy is written as [5]

$$E_c = A_{12}(1-\mathbf{m}_1\cdot\mathbf{m}_2) + 2B_{12}[1-(\mathbf{m}_1\cdot\mathbf{m}_2)^2]\,, \qquad (1)$$

where $\mathbf{m}_1$ and $\mathbf{m}_2$ are unit mean magnetization vectors of the two ferromagnets. The coefficients A_{12} and B_{12} are called *bilinear* (conventional exchange) and *biquadratic*, respectively. Biquadratic coupling in trilayers has recently been discovered by means of Kerr magneto-optic microscopy using specimens in which the thickness of the wedge-shaped chromium layer varies across the specimen [5]. They were deposited by molecular-beam epitaxy (MBE) on a (100)-GaAs-based substrate. At critical values of Cr thickness (near 0.6, 1.5, and 2.3 nm) the oscillating A_{12} vanishes and $B_{12}<0$ is revealed by the presence of static domains with 90° angles between $\mathbf{m}_1$ and $\mathbf{m}_2$. Such transition regions attributable to biquadratic exchange are also noted [6] in micrographs obtained by scanning-electron microscopy with polarization analysis (SEMPA) in wedge specimens composed of Fe/Cr deposited on a (100)-Fe whisker [7].

Here we address the question: Is the observed biquadratic coupling fundamental? In nonrelativistic n-electron quantum mechanics (neglecting the spin-orbit effect), the antisymmetry principle generally admits an isotropic effective coupling Hamiltonian for localized spin operators $\mathbf{S}_1$ and $\mathbf{S}_2$ (for an unambiguous example, between two Fe^{3+} ions in an insulator) which is expandable in the form $\sum_n J_n(\mathbf{S}_1\cdot\mathbf{S}_2)^n$. It would be very interesting to find macroscopic evidence reflecting something like the presence of the microscopic term $n=2$ in this fundamental Hamiltonian, or even something more general involving spin-orbit coupling, as suggested [5,8].

We will show, however, that the biquadratic coupling observed in Fe/Cr/Fe trilayers is in fact not of microscopic origin, but is still interesting for other reasons. The observed order of magnitude and negative sign for B_{12} arise phenomenologically from spatial fluctuations of microscopically *bilinear* coupling caused by terraced thickness fluctuations of the epitaxial Cr layer. Exchange stiffness of the ferromagnets tends to resist the torques from these fluctuations which induce static magnetic waves and thus relax the total energy, all of bilinear origin. Since these interlayer-mediated exchange torques exist in uniformly magnetized films only when $\mathbf{m}_1$ and $\mathbf{m}_2$ are not collinear, the energy relaxes only for $|\mathbf{m}_1\cdot\mathbf{m}_2|\neq 1$, leading to $B_{12}<0$ as observed. The biquadratic coupling is large when the interlayer is (100)-Cr because, as recently discovered, the short-period ($\simeq 2$ atomic monolayers) term in the bilinear coupling is large [7,8]. Our estimate of the short-period coupling required by the bilinear data agrees with one exchange theory [3,4] and independent experimental data. Thus biquadratic coupling has the significance that its measurement in specimens with structural defects can provide information about short-period exchange in ideal specimens.

Figure 1 shows a section of two infinite ferromagnetic films, having thicknesses D and D' and exchange stiffnesses A and A', separated by an interlayer of nonferromagnetic material. The corresponding magnetization vectors $\mathbf{M}$ and $\mathbf{M}'$, whose directions fluctuate about $\mathbf{m}_1$ and $\mathbf{m}_2$, respectively, are oriented in the x-y plane at angles $\theta(x,z)$ and $\theta'(x,z')$ measured from a common axis.

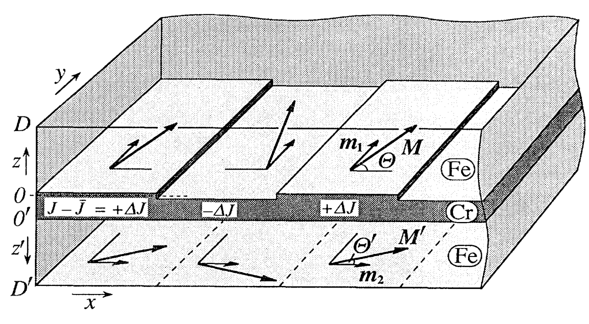

FIG. 1. Perspective section of an epitaxial trilayer magnetic film with periodic monolayer interfacial terraces. Arrows indicate schematically the continuous pattern of fluctuating static magnetizations $\mathbf{M}(x,z)$ and $\mathbf{M}'(x,z')$ in the x-y plane due to spatial fluctuations of exchange coupling $J(x)$ between the ferromagnetic Fe layers. (Special case $\mathbf{m}_1=\hat{\mathbf{x}}$, $\mathbf{m}_2=\hat{\mathbf{y}}$.)

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They are coupled to each other through the interlayer at $z=z'=0$. In the simplest version of our model, the conventional bilinear exchange coupling $J(x)$ per unit area is modulated in one dimension x with period $2L$ by fluctuations of interlayer thickness. These fluctuations consist of infinitely long monatomic growth terraces of width L separated by equal valleys on one interface, as indicated in Fig. 1.

Consider a single Fourier component $J_k \sin kx$ of this step function $J(x)$. The sum of exchange energies due to interlayer coupling and intralayer ferromagnetic stiffness per unit area is written

$$W_k=(2L)^{-1}\int_0^{2L} dx\{-J_k \sin kx \cos[\theta(x,0)-\theta'(x,0)] +A\int_0^D dz(\theta_x^2+\theta_z^2)+A'\int_0^{D'} dz'(\theta_x'^2+\theta_{z'}'^2)\}, \tag{2}$$

where subscripts x, z, and z' indicate partial derivatives. Here we have assumed that the exchange coupling acts between the inner-surface angles $\theta(x,0)$ and $\theta'(x,0)$ at common values of x (limit of very thin interlayer compared to L). We neglect the purely geometric aspect of the monolayer-scale interface roughness because it turns out that the significant length scales are L and D, which we assume are much greater than a lattice parameter. At the outer surfaces $z=D$ and $z'=D'$, the magnetizations are free.

By elementary variational calculus, setting $\delta W_k/\delta\theta=0$ leads to the static equilibrium conditions in the upper magnetic film:

$$\theta_{xx}+\theta_{zz}=0 \tag{3}$$

for all x and z, and

$$J_k \sin(\bar\theta-\bar\theta')\sin kx-2A\theta_z(x,0)=\theta_z(x,D)=0 \tag{4}$$

for all x. Because we regard J_k as small, we have replaced $\theta(x,0)$ and $\theta'(x,0)$ in $J_k\sin(\theta-\theta')$ with their means $\bar\theta$ and $\bar\theta'$. The system of equations (3),(4) has the solution

$$\theta=\bar\theta-\frac{J_k\sin(\bar\theta-\bar\theta')\sin kx\cosh k(D-z)}{2Ak\sinh kD}, \tag{5}$$

where the additive integration constant $\bar\theta$ contributes zero to the energy contained in fluctuations. The solution $\theta'(x,z')$ is given by a similar expression. These functions show how the vectors $\mathbf{M}$ and $\mathbf{M}'$ tend to draw together wherever $J>0$ and push apart wherever $J<0$, as indicated by the pattern of arrows in Fig. 1. Substituting these solutions into Eq. (2) and carrying out the integrals gives the static minimum $W_{k\,\min}$ of W_k.

Cases more general than Fig. 1 are represented by the two-dimensional Fourier series

$$J(x,y)=J_0+\sum_{\mathbf{k}\neq 0} J_{\mathbf{k}}\sin(k_x x+k_y y+\phi_{\mathbf{k}}), \tag{6}$$

where $\mathbf{k}=(k_x,k_y)$ and $\phi_{\mathbf{k}}$ is a phase angle, for exchange coupling across the x-y plane. The induced fluctuation pattern is given by superposition of wavelets, each similar to that in Eq. (5), directed in two dimensions. The general minimum energy works out to

$$W_{\min}=\sum_{\mathbf{k}\neq 0} W_{\mathbf{k}\,\min}=-8^{-1}\sum_{\mathbf{k}\neq 0}(J_{\mathbf{k}}^2/k)(A^{-1}\coth kD+A'^{-1}\coth kD')\sin^2(\bar\theta-\bar\theta'). \tag{7}$$

The summations in Eqs. (6) and (7) are carried over half of the $\mathbf{k}$ plane to avoid double counting of $\mathbf{k}$ and $-\mathbf{k}$. Since $\sin^2(\bar\theta-\bar\theta')=1-(\mathbf{m}_1\cdot\mathbf{m}_2)^2$, $W_{\min}$ contributes to the biquadratic term in Eq. (1). Note that the biquadratic coefficient is additive for the two ferromagnetic films, inasmuch as it represents the sum of energies contained within them. The sign of the predicted biquadratic coupling is predetermined ($B_{12}<0$, as observed) for any exchange distribution $J(x,y)$. In practice, the summation $\sum_{\mathbf{k}\neq 0}$ is usually dominated by one, or a very few, small $\mathbf{k}$ values.

To treat monolayer terraces, let us write for the thickness dependence of conventional exchange $J=A_{12}+(-1)^{n_{\mathrm{Cr}}}\Delta J$, where $A_{12}(n_{\mathrm{Cr}})$ is the long-period damped oscillatory coupling found by means of magnetoresistance [2] or $M(H)$ curves and Brillouin scattering of spin waves [8,9], n_{Cr} is the number of Cr monolayers, and $\Delta J(n_{\mathrm{Cr}})$ is the slowly varying amplitude of short-period (two atomic layers) coupling. For the monolayer terraces of width L shown in Fig. 1, we may write the periodic step function as

$$J(x)=A_{12}+\Delta J\,\mathrm{sgn}\sin(\pi x/L). \tag{8}$$

After Fourier analysis of this function, Eq. (7) works out to Eq. (1) with

$$B_{12}=\frac{-2(\Delta J)^2 L}{\pi^3 A}\sum_{m=1}^{\infty}\frac{\coth[\pi(2m-1)D/L]}{(2m-1)^3} \tag{9}$$

for equivalent ferromagnetic films. In this series, the first term alone suffices to good approximation because the remainder ($m\geq 2$) contributes less than 6% to the total. If the growth plane and (100) plane are slightly misaligned, both chromium faces have growth terraces. Then all terrace boundaries should be considered together in determining L, except that registered pairs are omitted as not presenting a change in J.

For experimental comparison, we assume the constant terrace width $L\simeq 10$ nm as estimated in order of magnitude from the spot profiles of low-energy electron diffraction (LEED) in the GaAs-based specimens [5]. Then the coth factors in Eq. (9) differ from 1 by less than 9% for the experimental Fe thicknesses $D=5$, 10, and 20 nm. This is consistent with the observation of nearly constant $B_{12}\simeq -0.13$ erg cm^{-2} [10] for these three specimens inferred from the breadth of the first transition region (exhibiting $\mathbf{m}_1$ perpendicular to $\mathbf{m}_2$ and centered near the mean chromium layer number $\bar{n}_{Cr}=3.5$) which lies between ferromagnetic ($\mathbf{m}_1=\mathbf{m}_2$) and antiferromagnetic ($\mathbf{m}_1=-\mathbf{m}_2$) regions created by the ramped chromium thickness. (However, we would predict a significant D dependence for L appreciably greater than these 10 nm.)

Altogether, three biquadratic coupling estimates, $B_{12} \simeq -0.13$, -0.009, and -0.008 erg cm^{-2}, are obtained at $\bar{n}_{Cr}=3.5$, 10.5, and 19, respectively [5]. Using Eq. (9) and the assumption $A \simeq 2\times10^{-6}$ erg cm^{-1}, we find the corresponding estimates $|\Delta J| \simeq 1.0$, 0.3, and 0.2 erg cm^{-2} for the short-period coupling amplitude in Eq. (8). In addition, very recent $M(H)$ studies [11] of similar specimens satisfying $4 \le \bar{n}_{Cr} \le 11$ reveal a maximum value of 0.2 erg cm^{-2} for $-B_{12}$ at $\bar{n}_{Cr} \simeq 4$, which would imply $|\Delta J| \simeq 1.2$ ergs cm^{-2} at this thickness. Thus the amplitude of short-period coupling required to explain biquadratic exchange ranges up to nearly the magnitude of the first negative maximum $A_{12}^{(1)} \simeq -1.4$ ergs cm^{-2} of the long-period coupling at $\bar{n}_{Cr} \simeq 5$ [9].

A two-parameter exchange calculation based on RKKY and superexchange terms [3,4] supports our interpretation. Its calculated long-period oscillations agree well with experiments [2,5,7–9]. The computed plots for the case of vanishing roughness [3,4] predict without further adjustment of parameters an additional exchange term with a period of two monolayers. The maximum of its slowly varying amplitude practically equals the magnitude of the first negative maximum of long-period oscillations. This prediction supports our above estimates from biquadratic coupling which satisfy $|\Delta J(n_{Cr})| < |A_{12}^{(1)}|$. In addition, two very recent direct measurements of short-period coupling support the order of magnitude of our estimates of $|\Delta J|$ [8,12,13].

It is significant that experiments [7,8,12,13] reflect remarkably well the predicted [3,4] zeros of long-period oscillation and the relative strengths of long- and short-period coupling terms discernible in the theoretical plots. Also, a one-layer "phase slip" of the domain reversal pattern in the range $\bar{n}_{Cr}=21$–24 is reported in an optimally ordered specimen [7]. In comparison, the theoretical plot shows a phase slip near $\bar{n}_{Cr}=36$ [4].

It is convenient to represent phenomenologically the kind of short-period term contained in the plotted exchange predictions for vanishing roughness [4] with the formula $\Delta J=\alpha\sin(\delta n_{Cr}+\phi)$, where δ is small and $\alpha(n_{Cr})$ is a monotonic slowly decreasing attenuation factor. Equation (7) or (9) then gives

$$|B_{12}| \propto \alpha^2(\bar{n}_{Cr})\sin^2(\delta\bar{n}_{Cr}+\phi)\,. \qquad (10)$$

Accordingly, B_{12} should nearly vanish at any phase-slip point $\bar{n}_{Cr}=n_{ps}$ satisfying $\delta n_{ps}+\phi=\pi s$, where s is an integer. This prediction is supported by the observation [6] that, in a highly ordered whisker-based specimen, the transitional bands of 90° orientation near the phase-slip position are narrower than neighboring such bands [14]. In particular, one could take $n_{ps}=24$, which is very near the center of a short-period antiferromagnetic region: The two nearest transitional bands (centered near $\bar{n}_{Cr}=23.5$ and 24.5) flanking this point appear decidedly narrower than the remaining ones displayed [14], as we would expect from the $\sin^2$ factor in relation (10). Although explained naturally by thickness fluctuations, this observation that B_{12} and ΔJ nearly vanish at the same chromium thickness presents a serious challenge to any more fundamental alternative explanation of the biquadratic effect.

The overall agreement between theory and experiment for both bilinear and biquadratic coupling arguably indicates that $\simeq$10-nm-wide monolayer terraces are the main structural defects affecting the exchange coupling of epitaxial Fe/Cr/Fe trilayers grown on GaAs. We note, by the way, very recent $M(H)$ measurements of two wedge-shaped trilayers with composition Fe/Al/Fe, which indicate that the exchange coupling varies gradually with Al thickness d_{Al} and that $|B_{12}| > |A_{12}|$ for 1.5 nm $< d_{Al} <$ 3.5 nm [15]. If this biquadratic coupling is again due to terraces, then we expect ideal specimens of Fe/Al/Fe to reveal that a short-period coupling, no trace of which has been observed directly, is dominant in this range of d_{Al}. In addition, very recent ferromagnetic resonance studies in trilayers using Cu interlayers reveal bilinear coupling described equivalently as an angle-dependent bilinear exchange [16,17]. Its observed sign and order of magnitude are consistent with our theory.

It is worth noting that Eq. (9) is inaccurate when the mean layer number $\bar{n}_{Cr}$ is near an integer and the Cr film is optimally uniform in thickness. For then the fraction of surface area occupied by assumedly compact mesas (or pits) is very small. This may well be the case in the best-ordered whisker-based specimens [7]. To treat this case, we adopt a model pattern, illustrated by the inset in Fig. 2, of square $a\times a$ mesas (or pits) centered on a square lattice of period P. Considering steps in $J(x,y)$

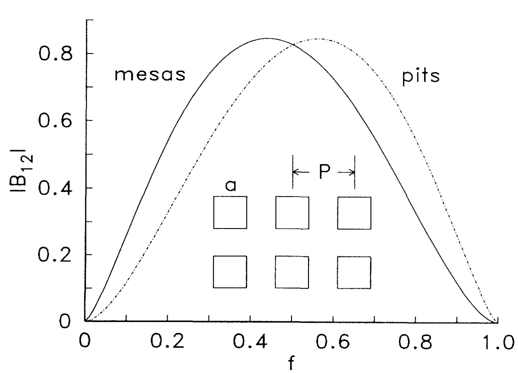

FIG. 2. Theoretical biquadratic coupling [Eqs. (7) and (11)] vs fractional terrace area f in one interface, assuming minimum possible terracing on a square lattice. The vertical scale is in units of formula (9) for $P=2L$.

equal to $2\Delta J$ at the edge of each mesa, we now find the Fourier coefficients in Eq. (6) to be

$$J_{\mathbf{k}} = \frac{4\Delta J}{\pi^2 n_x n_y} \sin(\pi n_x a/P) \sin(\pi n_y a/P) , \qquad (11)$$

with $\mathbf{k} = 2\pi(n_x, n_y)/P$. The vertical axis in Fig. 2 shows the biquadratic coupling, computed from Eqs. (7) and (11) for $A = A'$ and $D = D'$ in the limit of large D/P and normalized with respect to the infinite-mesa case of Fig. 1 and Eq. (9) with $P = 2L$. The horizontal axis shows the fractional mesa area $f = a^2/P^2$. Simply connected (e.g., square) mesas are more probable for $0 < f < \frac{1}{2}$, and simply connected pits (with $f = 1 - a^2/P^2$) for $\frac{1}{2} < f < 1$. Figure 2 shows how the biquadratic coupling in optimally structured specimens tends to zero whenever $\bar{n}_{\mathrm{Cr}} = n + f$ approaches an integer n or $n+1$. A consideration of this sort should be relevant to detailed interpretation of biquadratic coupling in the best whisker-based specimens [7].

Since demagnetization, neglected here, will generally diminish the amplitude of fluctuations, it weakens the biquadratic effect and our Eq. (7) is a lower bound of the energy correction. Extension of the theory to include demagnetization may be crucial for terrace widths significantly greater than the "exchange length" $A^{1/2}/M_s$ ($\simeq 7$ nm), which is close to the scale of interest as determined by terrace widths ($\simeq 10$ nm) in some GaAs-based specimens [5]. The anisotropic nature of dipolar interactions raises the possibility that demagnetization will lead to a more general coupling expression lacking the rotational invariance of Eq. (1).

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