Field-induced spin-reorientation transitions in magnetic multilayers with cubic anisotropy and biquadratic exchange

V. V. Kostyuchenko

Institute of Microelectronics, Russian Academy of Science, Universitetskaya 21, Yaroslavl, 150007, Russia

A. K. Zvezdin

Institute of General Physics, Russian Academy of Science, Vavilov 38, Moscow, 117942, Russia

(Received 7 May 1997)

A theoretical investigation of field-induced spin-reorientation phase transitions in magnetic multilayer structures is presented. We suppose that biquadratic and Heisenberg exchange energies between adjacent layers are of the same order of magnitude. The anisotropy energy is cubic. Our consideration is analogous to the Fe/Cr(100) superlattice system. We show that two phases with a noncollinear orientation of the magnetization in adjacent layers can represent the ground state. In our case an external magnetic field aligned perpendicular to the layers' direction induces spin-reorientation transitions in the plane of layers. A full list of such transitions is given. Other types of anisotropy are briefly discussed. [S0163-1829(98)03409-2]

I. INTRODUCTION

At the present time the exchange interaction of magnetic thin layers via a nonmagnetic spacer is the subject in a considerable amount of literature. It is well known that the main term of this interaction has a Heisenberg form $J_1\mathbf{n}_i\mathbf{n}_{i+1}$, where \mathbf{n}_i is a unit vector in the direction of magnetization in the *i*th magnetic layer. The value of J_1 is strongly dependent on the magnetic (and nonmagnetic) layers thicknesses and changes sign as the spacer thickness increases.^{1–3} A Heisenberg interaction tends to align the magnetization in adjacent layers parallel (for $J_1 < 0$) or antiparallel (for $J_1 > 0$). But it has been experimentally established that a noncollinear orientation of magnetization in adjacent layers is also possible.4-7 Therefore some other mechanism must be involved to account for the experimental data. For this reason a large number of different models for the non-Heisenberg interaction between adjacent layers is proposed. However, none of the theoretical models can quantitatively account for the experimental data.^{14,15} In most cases the non-Heisenberg exchange interaction is usually reported in terms of a biquadratic exchange form $J_2(\mathbf{n}_i \mathbf{n}_{i+1})^2$ (see Refs. 4 and 8–10) although other expressions are used also (see, e.g., Ref. 11).

In the case being considered in the zero anisotropy approximation phase transitions in magnetic multilayers are studied in Ref. 12. Nevertheless, the results obtained in Ref. 12 have only a qualitative meaning. In real multilayer magnetic materials the anisotropy energy has at least the same order of magnitude as the exchange energy. A uniaxial anisotropy in the plane of magnetic layers is taken into account in Ref. 13. But this paper discusses only phase transitions under the action of an external field parallel to the magnetic layers. We examine the case of a cubic anisotropy and biquadratic exchange. The case under consideration is analogous to the Fe/Cr(100) superlattice structure. In our paper we present some theoretical consequences of this model which can be verified rather easily, namely, the field-induced phase transition in magnetic multilayers.

According to the theoretical treatment and experimental observations J_2 is always positive. Thus biquadratic ex-

change tends to align the magnetization of adjacent layers in perpendicular directions and Heisenberg exchange in collinear directions. The joint action of the Heisenberg and biquadratic exchange brings into existence phases with a noncollinear orientation of the magnetization in neighboring layers. In the next section we examine the phase diagram for the spin-valve structure at zero field. Only four phases can satisfy the conditions of energy minimization and two of them are noncollinear.

As shown in Sec. III the action of an external magnetic field in the direction perpendicular to the layers gives rise to the renormalization of the exchange and anisotropy constants which determine the orientation of magnetization. Thus *perpendicular* to the plane the magnetic field induces changes of the magnetization orientation *in the plane* of layers.

II. ENERGY FUNCTIONAL AND PHASE DIAGRAM AT ZERO FIELD

Let us consider N magnetic layers mediated by N-1 spacer layers with an antiferromagnetic interaction between adjacent layers. In the case of an infinite layer number $N \ge 1$ or a spin-valve structure N=2 the energy functional has the form of a two-sublattice magnet.¹⁶ It differs from that of a classical antiferromagnet by the biquadratic exchange term. For definiteness sake we take the *z* axis perpendicular to the layer plane and the *x* axis along one of two easy axes in the layer plane.

The energy functional of that system can be written as

$$F = \sum_{i=1}^{2} \left[k \left[(n_i^x n_i^y)^2 + (n_i^z n_i^y)^2 + (n_i^x n_i^z)^2 \right] + \frac{m}{2} (n_i^z)^2 - h(n_i^z) \right] + \frac{1}{2} J_1(\mathbf{n}_i \mathbf{n}_{i+1}) + \frac{1}{2} J_2(\mathbf{n}_i \mathbf{n}_{i+1})^2.$$
(1)

Here J_1 is the Heisenberg exchange energy, J_2 the biquadratic exchange energy, k the energy of the cubic anisotropy,

5951

TABLE I. Properties of phases which minimize the energy functional (2).

	Phase	Stability condition	Energy
I	$\varphi_1 \!=\! \varphi_2 \!=\! 0$	$2k > J_1 + 2J_2$	$(J_1 + J_2)/2$
II	$arphi_1 \!=\! 0, \! arphi_2 \!=\! \pi$	$2k + J_1 > 2J_2$	$(J_2 - J_1)/2$
III	$\varphi_1 = -\varphi_2 = \frac{1}{2} \arccos \frac{J_1}{2(k-J_2)}$	$\sqrt{2} < \left \frac{J_1}{J_2 - k} \right < 2$	$\frac{J_1^2 - 4kJ_2 + 4k^2}{8(k - J_2)}$
IV	$\varphi_1 = -\frac{\pi}{2} - \varphi_2 = \frac{1}{2} \arcsin \frac{J_1}{2(k+J_2)}$	$\sqrt{2}(k+J_2) > J_1 $	$-J_1^2/8(k+J_2)$

h the Zeeman energy in an external magnetic field, and m the demagnetizing energy. All energies are measured in units of the magnetic field.

It is convenient to change the variables from rectangular \mathbf{n}_i to polar coordinates θ_i and φ_i , where θ_i measures the angle between the *z* axis and vector \mathbf{n}_i and the azimuthal angle φ_i measures the angle between the \mathbf{n}_i projection on the *x*-*y* plane and *x* axis. Thus the energy functional depends on four variables θ_1 , φ_1 , θ_2 , and φ_2 .

At zero external field a strong demagnetization field prevents any deviation of magnetization from the plane. Thus substituting in Eq. (1) $\theta_1 = \theta_2 = \pi/2$ we take

$$F = \frac{1}{2}J_1\cos(\varphi_1 - \varphi_2) + \frac{1}{2}J_2\cos^2(\varphi_1 - \varphi_2) + \frac{1}{4}k\sum_{i=1}^2\sin^2 2\varphi_i.$$
(2)

It is easy to verify that only four phases can satisfy the conditions for the global minimum of the functional (2). Two of them are collinear [ferromagnetic (phase I) and antiferromagnetic (phase II)] and two others are noncollinear with \mathbf{n}_1 and \mathbf{n}_2 symmetrical with respect to the easy axes (phase III) and hard axes (phase IV). The energy functional (2) has its minimum when a set of φ_1 , φ_2 is equal to one of four sets presented in the Table I. Minimum conditions and energy values for each set are displayed in Table I also. Figure 1 shows the phase diagram in the variables J_2/k and J_1/k .

This phase diagram may be interpreted as following. Due to the cubic anisotropy, phases I and II with $|\varphi_1 - \varphi_2| = 0, \pi$ and phase IV with $|\varphi_1 - \varphi_2| \simeq \pi/2$ have nearly the same anisotropy energy. Hence it follows that the ground state phase is determined by the J_1 and J_2 ratio. If it is granted that $|J_1| > J_2$, the collinear phase II (or I) is less energetic than the canted phase IV; otherwise, the phase IV is energetically preferable. It is interesting that for small anisotropy energy the transition from a collinear phase to a canted phase IV occurs via an intermediate phase, namely, the canted phase III with a nearly collinear orientation of the magnetization in adjacent layers.

III. FIELD-INDUCED SPIN-REORIENTATION TRANSITIONS

There is evidence that magnetization departs from the layer plane under the action of an external field directed in the perpendicular direction. As this takes place the problem of the energy functional (1) minimization becomes considerably more complex. It has not proved feasible to obtain an analytical solution of this problem in the general case. In actual conditions an exchange field and anisotropy field are far less than a demagnetization field. For the superlattice Fe/Cr, as an example, anisotropy and exchange fields are less than or equal to 1 kOe and the demagnetization field is on the order of 10–20 kOe. Then we can find from the equations $\partial F/\partial \theta_1 = 0$ and $\partial F/\partial \theta_2 = 0$ to a good approximation

$$\theta_1 = \theta_2 = \arccos(h/m).$$
 (3)

Substitution of these expressions into Eq. (1) gives an energy functional in the form of Eq. (2), where J_1 , J_2 , and k are replaced correspondingly by renormalized values

$$J_{1}(h) = \left(1 - \frac{h^{2}}{m^{2}}\right) \left(J_{1} + 2J_{2}\frac{h^{2}}{m^{2}}\right),$$
$$J_{2}(h) = \left(1 - \frac{h^{2}}{m^{2}}\right)^{2}, \quad k(h) = k \left(1 - \frac{h^{2}}{m^{2}}\right)^{2}.$$
(4)

By this means the equilibrium values of $\varphi_1(h)$ and $\varphi_2(h)$ can be defined by using the expressions in Table I, provided that instead of J_1 , J_2 , and k expressions (4) are taken. As $h \rightarrow m$, $k(h)/2J_1(h)$ and $J_2(h)/2J_1(h)$ tend to zero. Then



FIG. 1. Phase diagram in variables J_2/k and J_1/k . Roman numbering of phases is the same as in Table I. The solid line corresponds to the first-order phase transition and the dashed line corresponds to the second-order phase transitions.

according to the data in Table I, $|\varphi_1(h) - \varphi_2(h)| = 0, \pi$ as h = m. If $\varphi_1(m)$ and $\varphi_2(m)$ differ from $\varphi_1(0)$ and $\varphi_2(0)$, then the applied external magnetic field causes a spin-reorientation transition as h increased from 0 to m.

Investigation of field-induced phase transitions becomes easy to grasp through the use of the phase diagram in Fig. 1. The state of the multilayer system is completely determined by the two variables $\tilde{J}_1 = J_1(h)/k(h)$ and $\tilde{J}_2 = J_2(h)/k(h)$. Due to the condition $J_2(h)/k(h) = \text{const}$ [see Eq. (4)] the trajectory $\tilde{J}_2(\tilde{J}_1)$ has the form of a straight line parallel to the J_2 axis. As *h* is increased from 0 to *m*, \tilde{J}_1 changes from J_1/k to $+\infty$ for $J_1+2J_2>0$ and to $-\infty$ for $J_1+2J_2<0$. The intersection of this straight line and the lines of phase transitions in Fig. 1 shows possible field-induced phase transitions.

A full list of possible field-induced spin-reorientation phase transitions is given in the Appendix. It is interesting to note that the total number of field-induced transitions in some instances can be even 4 as h changes from 0 to m.

The most interesting case is $J_1 < 0$ and $J_2 > |J_1|/2$. Under these conditions $J_1(h) = 0$ at $h = h^* = m\sqrt{|J_1|/2J_2}$. Thus the effective Heisenberg exchange "disappears" at $h = h^*$ and the exchange field in the plane of layers becomes purely biquadratic. It is also vital to note that $J_1(h)$ changes sign at $h = h^*$.

The main conclusion $|\varphi_1(h) - \varphi_2(h)| \rightarrow \pi$ as $h \rightarrow \pi$ remains unchanged for other types of anisotropy (for $J_1 + 2J_2 > 0$). This fact can be explained as follows. If the angle between \mathbf{n}_i and \mathbf{n}_{i+1} exceeds $\pi/2$, then Heisenberg exchange acts as a repulsive force and biquadratic exchange as an attractive force. Otherwise both of these forces act as a repulsive force. Thus, if deviation of the magnetization from the basal plane exceeds $\pi/4$, then exchange forces tend to align the magnetization of adjacent layers so that the angle between the projection of magnetization in adjacent layers equals π . Our calculations for uniaxial anisotropy give support for this view.

IV. CONCLUSIONS

In this paper we have presented a theoretical investigation of the field-induced phase transitions in magnetic multilayers with cubic anisotropy and biquadratic exchange. The case of field perpendicular to the layer plane has been examined. The orientation of magnetization in the plane of layers is determined by the effective constants of anisotropy and exchange. The values of these constants are dependent on the value of the magnetic field. Thus a magnetic field perpendicular to the layers changes the orientation of the magnetization in the plane of layers.

In the particular case of ferromagnetic Heisenberg exchange between layers the value of the magnetic field can be chosen so that effective exchange interaction between magnetic layers becomes purely biquadratic. To the authors' minds experimental examination of these phase transitions can help to reveal the nature of the non-Heisenberg exchange interaction between layers.

ACKNOWLEDGMENT

The work was partially supported by the Russian Foundation for Fundamental Research (Grant 96-02-16250).

APPENDIX: FULL LIST OF FIELD-INDUCED PHASE TRANSITIONS (FIELD PERPENDICULAR TO THE PLANE OF MAGNETIC LAYERS)

There are no field-induced phase transitions if J_1 , J_2 , and k satisfy the following conditions:

- (1) $J_1 > 0$, $J_2 < 3k$, $2J_2 < J_1 k + \sqrt{k(k+2J_1)}$.
- (2) $J_1 > 0$, $J_2 > 3k$, $2J_2 < 2k + J_1$.

$$(3) \quad J_1 < 0, \quad J_1 + 2J_2 < 0.$$

Otherwise the following phase transitions occur as h changes from 0 to m:

(1)
$$J_2 < 3k$$
, $2J_2 > |J_1| - k + \sqrt{k(k+2|J_1|)}$.

First order at $h = h_1$.

(2)
$$J_2 > 3k$$
, $2J_2^2 > 2k^2 + J_1^2$.

- (a) First order at $h = h_2$.
- (b) Second order at $h = h_3$.

(3)
$$J_1 > 0$$
, $J_2 > 3k$, $2J_2 > 2k + J_1$, $2J_2^2 < 2k^2 + J_1^2$.

Second order at $h = h_3$.

(4)
$$J_1 < 0, \quad J_2 > 3k, \quad 2J_2^2 < 2k^2 + J_1^2, \quad J_1 + 2J_2 > k.$$

- (a) First order at $h = h_4$.
- (b) First order at $h = h_2$.
- (c) Second order at $h = h_3$.

(5)
$$J_1 < 0$$
, $J_2 > 3k$, $0 < J_1 + 2J_2 < k$

- (a) Second order at $h = h_5$.
- (b) First order at $h = h_4$.
- (c) First order at $h = h_2$.
- (d) Second order at $h = h_3$.

(6)
$$J_2 < 3k$$
, $2J_2 < -J_1 - k + \sqrt{k(k-2J_1)}$,

$$2J_2 + J_1 > 0, \quad J_1 < 0.$$

(a) First order at $h = h_6$.

(b) First order at $h = h_1$. Here the values h_i have the following meaning:

$$h_{1} = \frac{m}{\sqrt{2J_{2}(4J_{2}+3k)}} [2J_{2}(2J_{2}-J_{1}) + k(4J_{2}-J_{1}) - (J_{1}+2J_{2})\sqrt{k(k+J_{2})}]^{1/2}, \quad (A1)$$

$$h_{2} = \frac{m}{\sqrt{2(k^{2} + J_{2}^{2})}} [2(k^{2} - J_{2}^{2} - J_{1}J_{2}) + (J_{1} + 2J_{2})\sqrt{2(J_{2}^{2} - k^{2})}]^{1/2}, \quad (A2)$$

$$h_3 = m \left[\frac{2J_2 - 2k - J_1}{2(2J_2 - k)} \right]^{1/2}, \tag{A3}$$

$$h_{4} = \frac{m}{\sqrt{2(k^{2} + J_{2}^{2})}} [2(k^{2} - J_{2}^{2} - J_{1}J_{2}) - (J_{1} + 2J_{2})\sqrt{2(J_{2}^{2} - k^{2})}], \qquad (A4)$$

$$h_5 = m \left[\frac{2k - 2J_2 - J_1}{2k} \right]^{1/2}$$
, (A5)

$$h_6 = m \left[\frac{(J_1 + 2J_2)\sqrt{k(k+J_2)} - J_1k}{2J_2k} \right]^{1/2}.$$
 (A6)

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- ¹⁶Our investigations, aimed at finding structures with period T>2 layers, show that all these phases are metastable and so physically impracticable.