Magnetization processes and mechanism for kinetic anomalies in magnetic nanostructures

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Spin-reorientation phase transitions in anisotropic antiferromagnetic superlattices of the Co/Pt type are examined theoretically. Expressions are derived for the critical lines of second-order phase transitions to the ferromagnetic phase from the angular phase. Stability conditions are derived. They involve the external field, the anisotropic constant, the exchange energy, and the number of layers in the superlattice. A mechanism is proposed for explaining kinetic anomalies in superlattices. It incorporates a scattering of conduction electrons by "magnetic" energy barriers which exist if the magnetizations of neighboring magnetic layers are antiparallel. Expressions are derived for the mean free path and the magnetoresistance as a function of the magnitude of the external magnetic field. Numerical estimates are found.

1. Magnetic superlattices and multilayer structures of the Fe/Cr and Co/Cu type, which exhibit a giant magnetoresistance,[1–3] are attracting considerable interest. These new artificial materials and the giant magnetoresistance have been the subject of a large number of studies. Still, we lack a clear picture of the magnetization processes and the dependence of the critical fields on the number of layers, the surface anisotropy, the crystallographic or textural anisotropy, and the nature of the interlayer exchange. We believe that reaching an understanding of the magnetization processes in these materials, important in itself, is also of interest in connection with kinetic anomalies, one of which is the giant magnetoresistance. In particular, the magnetization reversal of the layers combines with some particular mechanism to determine the field dependence of kinetic effects. In this letter we are reporting a study of the phase diagram of a multilayer structure with an antiferromagnetic coupling between layers. To determine the relationship between the kinetic anomalies and the magnetic structure, we invoke a comparatively simple mechanism. This mechanism does not require the consideration of mechanisms for the scattering of conduction electrons accompanied by spin flip and/or the assumption that there is an important difference between the density of states for electrons with different spins in the corresponding layers. This mechanism incorporates a scattering of charge carriers by "magnetic" energy barriers which arise between the layers because of the antiparallel ordering of neighboring magnetic layers. This mechanism resembles in a certain sense the Andreev reflection of carriers at a boundary between normal and superconducting phases.[4] Again in this case we speak in terms of barriers which act

exclusively on electrons belonging to the subsystem with a certain spin. Since the total electron density is assumed to be the same in all the layers, barriers of this sort are not manifested as barriers of the electric potential.

2. The thermodynamic potential of an antiferromagnetic superlattice can be described at low temperatures by

$$F=-\sum_{i=1}^{N}\left(h\cos\vartheta_i-\frac{k}{2}\cos^2\vartheta_i\right)+\sum_{i=1}^{N-1}\cos(\vartheta_i-\vartheta_{i+1}), \tag{1}$$

where $h = dMH/\lambda M^2$ and $k = d\tilde{k}M^2/\lambda M^2$ are dimensionless parameters, d is the thickness of the magnetic layers, M is the magnetic moment per unit area of the atomic plane (in a layer of d planes), $\lambda > 0$ is proportional to the constant of the antiferromagnetic exchange interaction across paramagnetic layers, $\tilde{k}$ is the second-order anisotropy constant, ϑ_i are the angles made by the magnetizations of the layers with the field, and N is the total number of magnetic layers. Expression (1) is based on the assumption that the intralayer ferromagnetic interaction is much stronger than the interlayer antiferromagnetic interaction, so that the magnetization within each layer can be assumed to be uniform.

We first consider the case $k>0$, which corresponds to the case in which **H** is perpendicular to the easy axes. If $k>0$, the critical field for the transition to the ferromagnetic phase, h_{c2}, i.e., to a case in which the magnetizations of all layers are parallel to **H** ($\vartheta_i = 0$ for all ϑ_i), is found from the condition that the matrix of the second derivatives of the thermodynamic potential in (1) with respect to the variables ϑ_i and ϑ_k be positive definite.

If $\vartheta_i = 0$ for all ϑ_i, the matrix $A = \| \partial^2 F/\partial\vartheta_i\partial\vartheta_j \|$ has the elements

$$a_{11}=a_{NN}=h-k-1=b+1, \quad a_{ii}=h-k-2=b,$$

where $i=2, 3, \ldots, N-1$; $a_{i,i+1} = a_{i+1,i} = +1$; and all the other elements are zero. The condition under which all the characteristic numbers of this matrix are positive can be written in the form

$$\Delta_{N-1}+\Delta_{N-2}>1, \quad \Delta_{N-2}>0, \tag{2}$$

where Δ_k is the determinant of the matrix of rank k which is similar to matrix A but in which all the diagonal elements are the same, equal to b. Using the recurrence relation $\Delta_k^2 - \Delta_{k-1}\Delta_{k+1} = 1$, which can be proved by induction, and also using the lower estimate of b found from the second inequality in (2), $b>2\cos[\pi/(N-1)]$, we find $b>2\cos(\pi/N)$ or

$$h>h_{c2}=k+4\cos^2(\pi/2N). \tag{3}$$

When the magnetization of an antiferromagnetic crystal is perpendicular to the easy axes, the first critical field h_{c1} is zero.[5] In the case of a superlattice, this is not always true, since a superlattice with an odd number of magnetic layers will have a net ferromagnetic moment. At small values of k, and in the field interval $h_{c1} < h < h_{c1}^* < h_{c2}$, where h_{c1} and h_{c1}^* are certain fields, an antiferromagnetic configuration is stable (Fig. 1). As the number of layers is raised, the region in which this phase exists

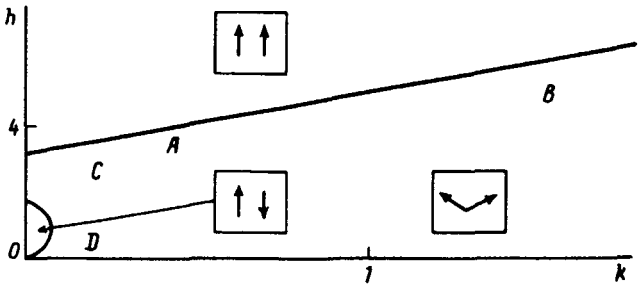

FIG. 1. Magnetic phase diagram of an antiferromagnetic superlattice with an odd number of magnetic layers. The field is perpendicular to the easy axes. The solid lines are lines of second-order phase transitions; line AB reflects expression (3), and line CD expression (6). Above AB, a ferromagnetic phase is stable. In the region bounded by curve CD, a phase in which the magnetization vectors are collinear with the field is stable, and an antiferromagnetic order is preserved. An angular phase exists elsewhere on this diagram.

shrinks, and in the limit $N \to \infty$ it vanishes. A study of the matrix A in this case can be reduced to a study of a matrix of rank $(N+1)/2$ of the type which we just discussed. In this case the diagonal elements are

$$b_{1,1}=b_{(N+1)/2,(N+1)/2}=(1+h-k)(2-h-k)-1=b^*, \tag{4}$$

$$b_{j,j}=(2-k-h)(2-k+h)-2=b, \quad j=2,3,\ldots,(N-1)/2.$$

In this case it is not possible to derive exact solutions, but by using the estimate $b>2\cos[2\pi/(N+1)]$, derived in the same way as above, and by using the relation $b \leqslant 2$, we can approximate the determinants $\Delta_k(b)$ as functions of b in the interval $2\cos[\pi/(k+1)] \leqslant b \leqslant 2$ by the linear law

$$\Delta_k(b) \frac{(k+1)[b-2\cos(\pi/(k+1))]}{4\sin^2[\pi/(2k+2)]}. \tag{5}$$

The approximation becomes exact at the boundaries of the interval. We can derive the following approximate ($N \gg 1$) condition:

$$k<\frac{h\left\{4\sin^2(\pi/(N-1))-\left[\frac{N+1}{N-1}\left(\frac{\sin[\pi/(N-1)]}{\sin[\pi/(N+1)]}\right)^2-1\right]h-h^2\right\}}{4\left\{\left[\frac{N+1}{N-1}\left(\frac{\sin[\pi/(N-1)]}{\sin[\pi/(N+1)]}\right)^2-1\right]+h\right\}}. \tag{6}$$

From this condition we can determine the maximum stability field (the condition $k=0$). At small values of k, the superlattice thus behaves as a ferromagnet. The orientation reversal occurs through two second-order phase transitions. Figure 1 shows the phase diagram of a superlattice with an odd number of layers.

3. When the field is parallel to the easy axes, the second critical field is given by expression (3) again, with $\mathbf{k}$ replaced by $-\mathbf{k}$, in the case in which the magnetization

process occurs through an angular phase in which the moments make various angles with the field. This situation requires an upper limit on $|\mathbf{k}|$: $|\mathbf{k}| < k^*$, where k^*, in the case of even N, for example, can be estimated from

$$\frac{4\cos^4(\pi/2N)-\sin^2(\pi/N-3)}{1+2\cos^2(\pi/2N)}<k^*<\frac{4\cos^4(\pi/2N)-\sin^2(\pi/N+3)}{1+2\cos^2(\pi/2N)}. \tag{7}$$

In the case $|\mathbf{k}| > k^*$, the transition to the ferromagnetic phase occurs through a first-order phase transition. Determining the nature of the overall magnetization process is a separate topic, lying outside the scope of the present letter.

In some cases, at certain layer thickness, particularly under the condition $\lambda \simeq 0$, we should consider an exchange interaction of higher order, whose energy can be described by the phenomenological expression

$$F_{ex}' = \sum_{i=1}^{N-1} \frac{\lambda'}{\lambda} M^2 \cos^2(\vartheta_i - \vartheta_{i+1}). \tag{8}$$

Here $\lambda' > 0$ is the biquadratic exchange constant. This expression should be added to thermodynamic potential (1). The existence of this contribution, which was first introduced by Ruhrig *et al.*,[6] can be explained in terms of (for example) fluctuations in the thickness of the nonmagnetic spacer layers.[7] The expressions for the second critical field remain similar to (3), but with renormalized parameters h and k, which should be divided by $(1+2\lambda'/\lambda M^2)$.

4. We turn now to the nature of the motion of electrons in multilayer structures of this sort. We introduce a quasichemical potential for electrons with a given spin:

$$\zeta_\sigma = \zeta + \sigma\Delta(\mathbf{r}), \tag{9}$$

where $\Delta(\mathbf{r})$ is the Zeeman energy due to the effect of the external and internal exchange fields, $\sigma = \pm 1$ is the index of the electron subsystem ($\downarrow$ or $\uparrow$), and ζ is the chemical potential. The potential $\zeta_i(\mathbf{r})$ is a periodic function with a period equal to twice the period of the superlattice, $2D = 2(d + d_2)$, where d_2 is the thickness of the layers of the nonmagnetic component of the superlattice. The function $\zeta_\uparrow(\mathbf{r})$ is shifted by half a period. We wish to stress that although the change in $\zeta_\sigma(\mathbf{r})$ is correlated with the change in the crystal structure, it is associated with the distribution of the magnetization of the system, $M(\mathbf{r})$, which is determined by exchange effects. The height U of the barrier separating neighboring wells in the $\zeta_\sigma(\mathbf{r})$ potential well is determined by the relative orientation of the magnetizations of neighboring layers. It vanishes in the ferromagnetic phase. Ignoring contact effects at the interface for simplicity, we consider elastic collisions of electrons with the barriers. The change in the quasimomentum $p = mv = \hbar k$ can be estimated from

$$\delta p \approx (-dU/dz) L_{ex}/v, \tag{10}$$

where $(-dU/dz)$ is the force exerted on an electron by the barrier, which can be estimated as $(-dU/dz) \approx U/L_{ex}$, L_{ex} is the characteristic decay length of the induced spin polarization in the nonmagnetic metal, U is the barrier height ($U = 2\Delta_0$ for $h=0$; $U=0$ for $h > h_{c2}$), and $v \approx v_F$ is the electron velocity (which is on the order of the Fermi velocity). Substituting these estimates into (10), we find

$$\delta p \approx U/v \approx p_F U/E_F \ll p_F, \tag{11}$$

where p_F and E_F are the Fermi momentum and energy. The change in the quasimomentum thus turns out to be small in comparison with the quasimomentum itself. This is a typical situation for a scattering by spatial irregularities which vary slowly (in comparison with the wavelength). Since the length scale of the irregularities is shorter than the mean free path determined by scattering by impurities and lattice defects, the scattering process described above is pertinent.

It is natural to suggest that the scattering is not specular, because of the obvious roughness of the barriers, and that the scattering can be described with the help of a specularity parameter p^*.

On the basis of the arguments above and (11), one might suggest that as electrons with a certain spin interact with the barrier, a part of the momentum equal to $p = 1 - U/E_F$ is conserved, while a part equal to U/E_F undergoes a complete randomization (as a result of a diffuse scattering by the rough boundary). If we use a model of scattering in thin platelets, we should thus set the specularity parameter at $p^* = 1 - U/E_F$. The expression for the effective mean free path Λ_{mw} in the angular phase at $h < h_{c2}$ is

$$\frac{\Lambda_{mw}}{\Lambda_0} = \frac{3}{4\pi S_c} \int\int \left\{ 1 - \frac{(1-p^*)\exp(-|\mathbf{r}-\mathbf{r}_B|/\Lambda_0)}{1-p^*\exp(-|\mathbf{r}_B-\mathbf{r}_B'|/\Lambda_0)} \right\} \cos^2\vartheta \, d\Omega \, dS_s, \tag{12}$$

where $d\Omega$ is an element of solid angle containing the vector $\mathbf{r} - \mathbf{r}_B$, ϑ is the angle between $\mathbf{r} - \mathbf{r}_B$ and the electric field gradient, dS_c is a small area perpendicular to the field gradient, S_c is the cross-sectional area, $\mathbf{r}_B'$ is the starting point on the surface for a particle which was subsequently reflected specularly in the v direction, and Λ_0 is the mean free path in the ferromagnetic phase.[8] In the Knudsen limit $\Lambda_0 \gg D$, this integral simplifies:[9]

$$\frac{\Lambda_{\text{eff}}}{\Lambda_0} \simeq \frac{3}{8} \frac{D}{\Lambda_0} \frac{1+p^*}{1-p^*} \ln\left(\frac{2\Lambda_0}{D}\right). \tag{13}$$

Substituting (13) into the expression[8]

$$\sigma_\alpha = e^2 \Sigma \Lambda_\alpha / (12\pi^3 \hbar) \tag{14}$$

for the electrical conductivity (Λ_α is the mean free path in the α phase, and Σ is the area of the Fermi surface in k-space), and assuming $p^* = 1 - U/E_F$, we can easily show that the giant magnetoresistance $(R_{af} - R_f)/R_f = \Delta R/R$, where R_{af} and R_f are the resistivities of the antiferromagnetic phase ($h < h_{c2}$) and the ferromagnetic phase ($h > h_{c2}$), is given by

$$\frac{\Delta R}{R} = \frac{\Lambda_0}{\Lambda_{mw}} \simeq \frac{8}{3} \frac{\Lambda_0}{D} \frac{U(h)}{E_F} \left[\ln\left(\frac{2\Lambda_0}{D}\right) \right]^{-1}. \tag{15}$$

Under typical conditions we would have $\Lambda_0/D \approx 5$ and $U/E_F \approx 0.1$ and thus $\Delta R/R \approx 0.5$.

Expression (15) can be used to derive the field dependence of kinetic effects which depend on the mean free path, since the barrier height is a function of the

external field. To do this we need to numerically solve the equation for an extremum of thermodynamic potential (1), derive expressions for the barrier height, and average the resulting expressions over the entire superlattice, since the orientation of the magnetization in the superlattice is not uniform. If the number of layers is sufficiently large, we can use the following expression, which ignores the nonuniformity of the magnetization:

$$U(h)=2\Delta=2\Delta_0\sin(\psi)=2\Delta_0[1-(h/h_{c2})^2]^{1/2}, \tag{16}$$

where ψ is the angle made by the magnetization with the external field, and h_{c2} is given by expression (3) or analogs thereof. This expression, along with (15), describes the field dependence of the giant magnetoresistance.

Working from the model discussed above, we can, in a very natural way, predict changes in other kinetic characteristics which depend on the mean free path (e.g., thermoelectric and galvanomagnetic effects, the thermal conductivity, and rf and optical effects) in the course of a phase transition from an antiferromagnetic state to a ferromagnetic one in multilayer films and superlattices of this sort. A planar geometry is not particularly important here. This behavior is characteristic of a wide range of composite systems (Ref. 10, for example) with an antiferromagnetic interaction between nearest elements of the system which can be suppressed by an external magnetic field.

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