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Spin scattering in ferromagnetic thin films

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By combining parallel and transverse magnetoresistance measurements on thin films of Co and Ni, the contribution of spin scattering at the domain walls is separated from the anisotropic magnetoresistance (AMR). A model, based on the Larmor-precession-induced deviation of the conduction electron spin direction during domain-wall traversal is developed. By using a scattering probability which varies with the cosine of the angle between the carrier spin and the local exchange field (as used for giant magnetoresistance systems) it is possible to account for the amplitude of the measured magnetoresistive effect.

Giant magnetoresistance (GMR) is a popular research topic owing to the theoretical challenges it poses and the promise of new technological devices. GMR arises in a variety of magnetic systems including heterostructures and multilayers,[1] spin valves,[2] or granular materials,[4] whose common property is that they consist of separated regions of magnetic material capable of adopting independent magnetization orientations in response to externally applied magnetic fields. The effect has its origin in spin-dependent momentum scattering of the conduction electrons which may be controlled by changing the magnetic configuration. Viewed differently, the GMR arises from the transfer of magnetic information from one magnetic region to another by the carriers across the intermediate nonmagnetic spacer metal, and the size of the effect depends on the efficacity with which the information is coded on the chemical potentials of up and down spin channels, and also on the decay of the coding in transit.

This interpretation of GMR in heterostructures begs the question as to why analogous spin-scattering effects are not observed at interfaces between differently magnetized domains in pure ferromagnets, which simply correspond to GMR trilayer systems with the central nonmagnetic layer replaced by the domain wall.

The ordinary magnetoresistance of pure ferromagnetic materials as a function of applied magnetic field and temperature has been intensively studied in the 1960s and 1970s.[5–8] It consists of two components: a contribution from the bulk ferromagnet, part of which is the anisotropic magnetoresistance (AMR),[5,9] and a smaller contribution from the domain walls. These may be disentangled as described below.

Owing to its small size and the consequent difficulty in measurement, scattering by domain walls has received sparse attention; however, two views can be found in the literature. Berger[7] proposes that, because the conduction electron wavelength is much shorter than the domain-wall width, the electronic spin follows the local magnetization adiabatically and gradually tilts as it traverses the wall. Cabrera and Falicov[8] have treated the problem of domain-wall-induced electrical resistivity in Fe analytically by examining the difference in reflection coefficient at a domain wall for up and down spin electrons. The domain wall essentially presents a potential barrier whose height is different for the two-spin channels owing to the exchange field.

Here we report room-temperature magnetoresistance measurements on cobalt and nickel films which lead us to propose a new model based on the pseudo-Larmor precession of the electron spin about the changing exchange field direction in the wall. The dissipative mechanism invoked is identical to that use to model GMR in that the probability of carrier scattering varies with the cosine of the angle between the spin and the exchange field.[4] This angle arises from the nonadiabaticity of the spin's passage through the domain wall.

The thin ferromagnetic films were deposited by laser chemical Vapor deposition (LCVD) or pulse laser deposition (PLD). The results presented here were obtained on two 5-mm-square samples: a Co film deposited by LCVD on glass, consists of grains of hcp cobalt randomly oriented with typical grain size of 200 nm (as measured on SEM and AFM micrographs). The film thickness was measured by Rutherford backscattering spectrometry to be 28 nm. A Ni film, made by PLD on glass, is about 30 nm thick and a typical crystallite size of 20 nm was inferred from x-ray diffraction. The residual resistivities were measured to be 12 $\mu\Omega$ cm for the Co film and 17 $\mu\Omega$ cm for the Ni film, values which are typical for nanocrystallite metals. Hysteresis curves with the applied field B_0 in plane were measured by transverse Kerr effect and also by vibrating sample magnetometry. The coercive fields inferred from the Kerr loops are $\mu_0 H_c(\mathrm{Co})=4.5$ mT and $\mu_0 H_c(\mathrm{Ni})=17$ mT.

The measurements of the magnetoresistance as function of the external field were carried out with a low-frequency in-plane current density of 2.10^5 A/m^2. The curves obtained, which are presented in Fig. 1, show a small effect of order 0.2%. As expected, because the atomic charge distributions of both Co and Ni are oblate, the resistance is higher when the current is parallel to the magnetization and lower in the transverse geometry. This reflects the effect of spin-orbit coupling and is consistent with measurements carried out on bulk ferromagnetic samples.[5] The minima obtained in the longitudinal geometry and the maxima in the transverse geometry correspond to the maximum disorder of the distribu-

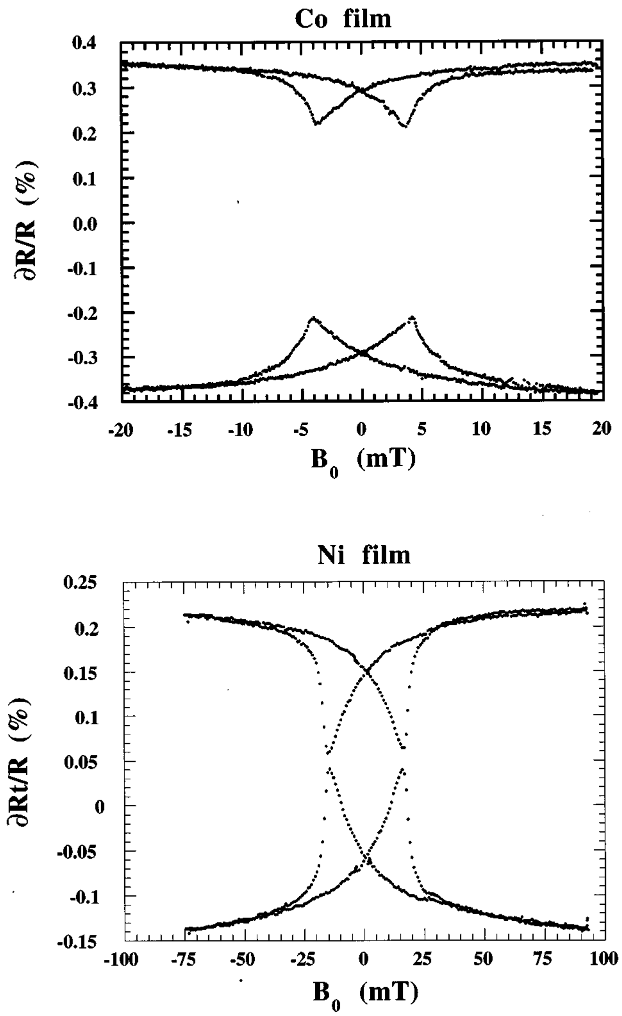

FIG. 1. Magnetoresistance measurements in longitudinal ($\rho_\parallel$) and transverse ($\rho_\perp$) geometries for Co and Ni. The peaks occur at $\pm\mu_0 H_c$.

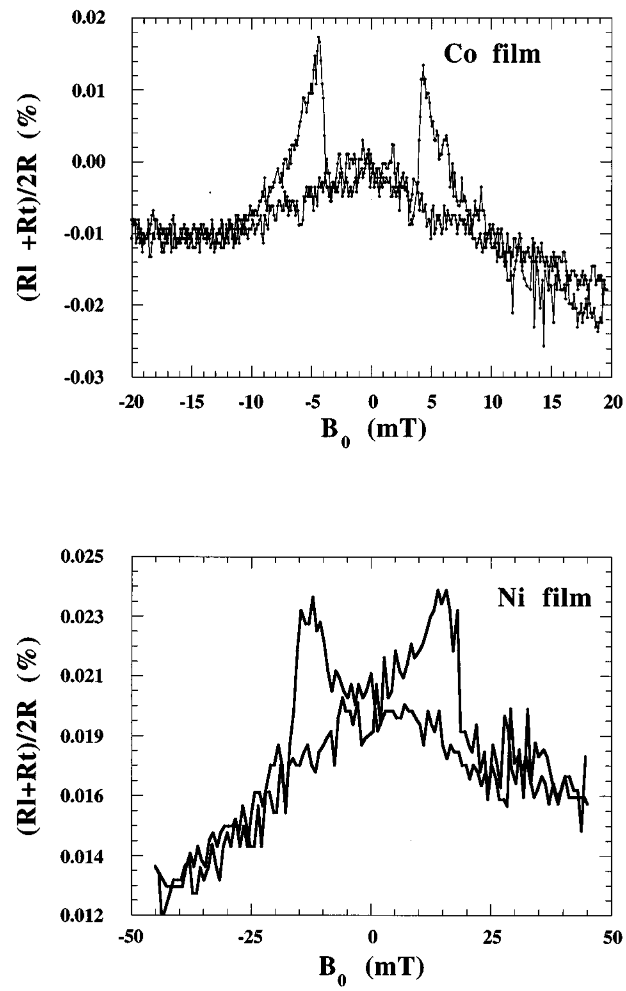

FIG. 2. Domain-wall-scattering induced resistivity obtained by adding transverse and longitudinal magnetoresistance curves.

tion of magnetic domains at the coercive field. It is well known[9] that the anisotropic change of resistivity (AMR) of ferromagnetic materials as function of θ, the angle between the local magnetization and the current lines, can be expressed as

$$\rho=\rho_\perp+(\rho_\parallel-\rho_\perp)\cos^2\theta$$

with $\rho_\parallel$ and $\rho_\perp$ the resistivities with the current parallel or perpendicular to the magnetization. When the material is composed of several magnetic domains, all the local magnetizations contribute to the total resistivity. In fact, we can notice that if we rotate the current by 90°, each magnetic domain will have its resistance changed following the 90° rotation of the atomic scattering cross section to become $\rho=\rho_\perp+(\rho_\parallel-\rho_\perp)\cos^2(\theta+90°)=\rho_\perp+(\rho_\parallel-\rho_\perp)\sin^2\theta$. If the current density is completely homogeneous and the resistive contributions additive (which is the case when the more resistive regions are isotropically distributed throughout the sample), we obtain a constant value for the sum of the two global resistivities (longitudinal and transverse). This result is useful because by adding the two resistivity curves in longitudinal and transverse geometry, the angular contribution of galvanomagnetic effects are eliminated. In fact, granular magnetic materials where the current density is homogeneous are ideal systems for this operation.[10] In our case however, the magnetic domains have stripelike shapes which prevent the current from being strictly homogeneous. This effect leads to a decrease in the resistance which can be estimated by a simple calculation of two-channel conduction in parallel.[11]

The average $(\Delta\rho_l+\Delta\rho_t)/2\rho_l$ for Co and Ni are presented in Fig. 2 where positive humps appear at the coercive field which have an amplitude of order 10% of the AMR effects. For both Co and Ni the measurements show an excess resistivity at $\pm H_c$ about 10 times larger than the expected (negative) error on the galvanomagnetic effects.[11] We attribute this extra resistance to spin scattering of the conduction electrons at the border between magnetic domains, through the same mechanism as the one responsible for the GMR effect. To our knowledge, this is the first time the spin-scattering contribution to the magnetoresistance of pure ferromagnetic materials has been extracted from AMR data.

Two key length scales in the problem of magnetoresistance in magnetic structures are λ_θ, the mean free path (mfp) of an electron whose spin s makes an angle θ_s with the local magnetization direction, and λ_s the spin diffusion length which is the average distance an electron can travel before it encounters a scattering event that flips its spin into a state where it is parallel to the local magnetization. In ferromagnets, the momentum scattering rate of polarized electrons is a linear function of the cosine of the angular deviation between the electron spin and the scattering center local magnetic moment, $\cos(\theta_s)$.[4] The mfp can be expressed as

$$\lambda_\theta = \frac{\overline{\lambda}}{1+p^2+2p\cos(\theta_s)}$$

with $\overline{\lambda}$ the average mfp and p the spin-dependent scattering ratio. Any angular deviation between conduction electrons spins and local magnetization should give rise to an extra resistivity given by

$$\frac{\Delta R_w}{R_w} = \frac{2p}{(1-p)^2}(1-\langle\cos\vartheta\rangle) \quad \text{per domain wall.}$$

In total, the magnetoresistance of the sample can be expressed as a function of the magnetic domain size d_s and the wall width δ_w as

$$\frac{\Delta R}{R} = \frac{2p}{(1-p)^2}(1-\langle\cos\vartheta\rangle)\frac{\delta_w}{d_s}.$$

We propose to apply this result to the problem of domain-wall crossing by conduction electrons where the average angle between electrons spins and local moment can be estimated.

Cabrera and Falicov[8] studied the extra resistivity induced by domain-wall crossing by calculating the transmission and reflection coefficients of electrons tunneling through walls in two limiting cases: small band splitting with 180° wall of arbitrary width, and large band splitting with 180° narrow wall. However, for strong ferromagnets like Co and Ni, the splitting cannot be considered small, and the second approach is valid only in the "sudden" approximation in which the effective domain-wall magnetization rotation frequency: $w_{\mathrm{wall}} = \nu_{\mathrm{Fermi}}/2d_w$ is much larger than the Larmor precession frequency of the carrier spin: $\upsilon_{\mathrm{Larmor}} = E_{\mathrm{exchange}}/h$. As seen from considerations of the Jitterburg spin-mixing mechanism[12] in superparamagnetic particles, the Larmor precession translated into spatial frequency has a wavelength comparable to the diameter of the superparamagnetic particles (about 30 Å) and hence is less than a typical domain-wall width. Thus, the electron spin traverse of a domain wall is an intermediate case between the "sudden" and the "adiabatic rapid passage" approximations of magnetic resonance. The electron spin attempts to follow the changing local magnetization direction as it traverses the wall, but because the pseudo-Larmor frequency is not sufficiently high to make the process perfectly adiabatic, the spin deviates from the magnetization vector in passing. Figure 3 shows graphically the results of a numerical simulation of the domain-wall process in which the average cosine of the deviation can be estimated considering that the electron spin catches back onto the local spin after one turn at the Larmor frequency.

The average cosine of the angle between electron spin and local moment in the wall is the projection of the cone of angle θ_0 made by the electron spin on one of its sides:

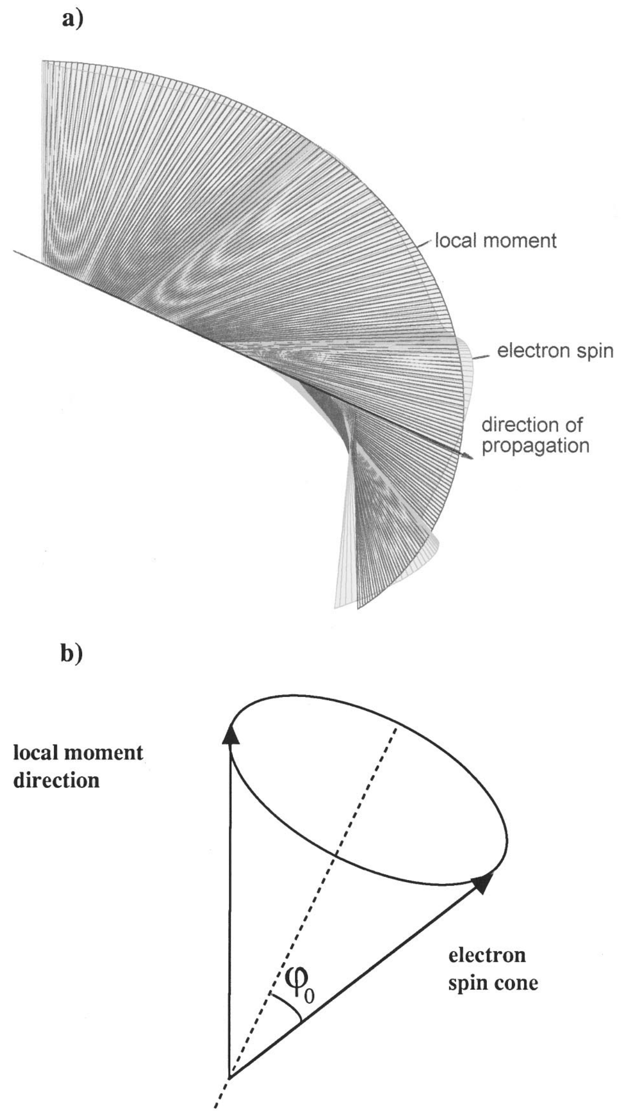

FIG. 3. Numerical simulation of the canting of the conduction electron spin as it attempts to follow the local magnetization during transversal of the domain wall: (a) in the laboratory frame of reference, (b) in the frame of reference of the local moment.

$$\langle\cos\vartheta\rangle = \cos^2\vartheta_0.$$

The angle θ_0 can be estimated as the angle the local moment rotates during half a Larmor precession (Fig. 3):

$$\vartheta_0 = \frac{1}{2}\left(\frac{\pi}{(\delta_w/\nu_{F\perp})\nu_{\mathrm{Larmor}}}\right) = \frac{\pi h \nu F_\perp}{2\delta_w E_{\mathrm{ex}}}$$

with $\nu_{F\perp}$ the component of the Fermi velocity perpendicular to the wall (the drift velocity due to the applied electric field being neglected). By averaging over the Fermi surface (considered spherical), we get, for the average angle of conduction electrons,

$$\vartheta_0 = \frac{h\nu_F}{\delta_w E_{\mathrm{ex}}}.$$

And for small angles where $\sin\vartheta_0 = \vartheta_0$ and by substituting it in the expression for the extra resistivity we get the final expression for the magnetoresistance:

$$\frac{\Delta R}{R}=\frac{2p}{(1-p)^2}\left(\frac{h\nu_F}{E_{\rm ex}}\right)^2\frac{1}{\delta_w d_s}.$$

The quantity p represents the asymmetry of spin scattering in ferromagnetic materials. The ratio of the mean free paths for spin-up and spin-down electrons have recently been measured in materials of a quality comparable to ours.[3,13] The values measured for Co and Ni are close to 5, giving for the quantity $2p/(1-p)^2$, values of order of unity.

From consideration of the residual resistivity of the sample it may be seen that the mean free paths of the electrons are small compared with the domain-wall thickness. Consequently, the simple picture of a spin which makes a totally ballistic traverse of the entire domain wall needs some modification, and the spin trajectory represents a diffusive process (which comprises many short paths each making a different angle with the wall direction) on which is superimposed the drift motion induced by the electric driving field. However, the essential point is that the mean free path is large enough so that the spin of the electrons traveling perpendicular to the domain wall experience an average angle deviation similar to that of ballistic electrons. The relevant quantity to be compared to the mean free path is half of the Larmor period [see Fig. 3(b)]. As long as this distance is exceeded by each electron traveling perpendicular to the wall, each will experience the full effect of the angular deviation during the time they spend in that direction. After scattering, the change of local moment canting viewed by the electron with a momentum direction not perpendicular to the wall is much slower, and its effect is strongly reduced. However, at any time, the number of electrons traveling perpendicular to the wall remains constant, and the total effect of the spin-angle deviation is the same as for purely ballistic traversing of the wall.

The large resistivities ($\rho=19$ $\mu\Omega$cm for Co and $\rho=26$ $\mu\Omega$cm for Ni) measured in our films are caused by a combination of defect scattering in the bulk (dislocations, impurities, strain) and grain boundary scattering. By direct comparison with measurements on films of comparable quality[3,13] we estimate the majority electron mean free path of our films to be of the order of 40 Å. This value is larger than the half Larmor periods of about 15 Å in Co and 30 Å in Ni and the calculation for ballistic electrons applies to our films.

It is well known that the character of the magnetic order observed in domain walls varies from system to system and is distinctive in thin films.[14] For simplicity of the calculation, we assumed a canting that is linear with the distance in the wall. It should be noted that our model is relatively insensitive to these variations and relies only on a relatively smooth variation of magnetization direction with distance across the domain wall. We checked in our numerical simulations that the effect of the nonlinearity of pure Bloch and Néel walls is small.

It is evident in our final expression that thinner domain walls make a more significant contribution to the magnetoresistance. In fact, the thinner the domain wall, the more the system approximates to that of a GMR structure in which the spin passage is truly "sudden." In the materials discussed here, we are in both cases close to the adiabatic approximation but the carriers spin experience more or less difficulty in following adiabatically the changing orientation of the local exchange field.

Magnetic domain sizes have been measured by decorating the walls with an oil-based ferrofluid. The average domain widths measured on optical micrographs are $d_{s\mathrm{Co}}=4$ μm and $d_{s\mathrm{Ni}}=50$ μm (both much greater than the grain size). By using 15 nm for the average domain wall width of Co and 100 nm for Ni and by taking typical exchange energies[15] of 1 eV for Co and 0.3 eV for Ni, the model gives for the spin-scattering magnetoresistances

$$\left.\frac{\Delta R}{R}\right|_{\rm Co}=3\times10^{-4}\quad\text{and}\quad\left.\frac{\Delta R}{R}\right|_{\rm Ni}=4\times10^{-5},$$

which compare to the experimental values of

$$\left.\frac{\Delta R}{R}\right|_{\rm Co\ expt}=2\times10^{-4}\quad\text{and}\quad\left.\frac{\Delta R}{R}\right|_{\rm Ni\ expt}=3\times10^{-5}.$$

In conclusion, we have demonstrated that precise measurement of the magnetoresistance permits the separation of the spin-scattering contribution from the galvanomagnetic effects in thin films of cobalt and nickel. A model which explains the measured extra magnetoresistance is based on the Larmor precession of conduction electrons spins which attempt to follow the local magnetization while crossing domain walls. The process is not purely adiabatic and the mean free path of conduction electrons is reduced according to the average cosine of the angle between electron spin and the local magnetization. The calculated values for the extra resistance generated by domain-wall transversal are in very good agreement with the experimental results on both Co and Ni films. It is worth pointing out that it should be possible to increase the magnetoresistive effect of the domain walls by making them thinner or denser. This can be controlled by adjusting the thickness of the films[14] and by using the magnetocrystalline anisotropies (especially for Co) in some particular geometry. The nature of the walls and their density could then be monitored. In particular, crosstie walls or ripples in the magnetic configuration are expected to enhance the magnetoresistance.

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[11] In the most unfavorable case, two categories of domains with their local magnetization either parallel or perpendicular to the current lines form stripes running along the entire sample length. These two conducting channels are seen in series by a longitudinal current and in parallel by a transverse current. If we call ρ_t the resistivity measured in transverse geometry (magnetic domains in series) and ρ_l that measured in longitudinal geometry (domains seen in parallel), we get $\rho_t + \rho_l = \rho_\parallel(1-x) + \rho_\perp/(1-x)$ with $x = 1-\rho_t/\rho_\parallel$ being a small quantity.

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