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Interfacial roughness and angle dependence of giant magnetoresistance in magnetic superlattices

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Abstract. Using the two-point conductivity formula, we numerically evaluate the giant magnetoresistance (GMR) in magnetic superlattices with currents in the plane of the layers (CIP), from which the effect of the interfacial roughness and magnetization configuration on the GMR is studied. With increasing interfacial roughness, the maximal GMR ratio is found to first increase and then decrease, exhibiting a peak at an optimum strength of interfacial roughness. For systems composed of relatively thick layers, the GMR is approximately proportional to $\cos^2\theta$, where 2θ is the angle between the magnetizations in two successive ferromagnetic layers, but noticeable departures from this dependence are found when the layers become sufficiently thin.

PACS. 75.70.Pa Giant magnetoresistance – 75.70.-i Magnetic films and multilayers – 72.10.Bg General formulation of transport theory – 72.15.Gd Galvanomagnetic and other magnetotransport effects

Giant magnetoresistance (GMR) observed in magnetic multilayers with antiferromagnetic coupling between successive ferromagnetic (FM) layers [1] has attracted considerable theoretical and experimental interest because of its potential applications. The resistivity of such a magnetic layered structure can drop tens percent when an applied magnetic field is used to overcome the antiferromagnetic coupling, leaving the structure in a state where the magnetic moments of all the FM layers are aligned. It is widely accepted that this novel transport phenomenon arises from the spin-asymmetric scattering at the interfaces between the ferromagnetic (FM) and nonmagnetic (NM) layers and in the bulk of the FM layers. Although there still exists controversy whether the spin-dependent bulk scattering or interfacial scattering dominates the GMR behavior, the importance of the spin-dependent interfacial scattering to the GMR has been stressed in both experimental [2–4] and theoretical [5–7] works. There are two important factors of the interfacial scattering: its strength and its spin dependence. That scattering at the interfaces is stronger than in the bulk of the layers is generally acknowledged, because there is a rapid change in the electronic structure in the interfacial regions. Recently, it has been shown [8] that correlation in the scattering from pairs of interdiffusive atoms at the interfaces may lead to constructive interference between scattering amplitudes, and hence enhance the spin dependence of the scattering at the interfaces. It is therefore of extensive interest to carry out detailed study on the effects of the enhanced spin-dependent interfacial scattering on the GMR.

On the experimental side, the strength of the interfacial scattering can be adjusted by varying interfacial roughness. Fullerton *et al.* [2] varied the interfacial roughness of sputtered Fe/Cr superlattices by three independent methods; changing sputtering gas pressure, varying sputtering power and increasing the total thickness of the superlattice. They reported that the GMR was substantially enhanced by increasing the interfacial roughness. However, the opposite result was found as well that increased interfacial roughness decreases the GMR [3,4]. In the experiment of Parkin [3], the interfacial roughness was increased by annealing the sample at elevated temperatures, which causes increased dissolution of the FM/NM interlayers, and a decreased GMR was found. Thomson, Riedi and Greig [4] showed that the Co/Cu (111) multilayers grown by MBE had an increase in GMR as the interfaces became more abrupt on an atomic scale. These experiments look to have given contradictory results, so that a reasonable theoretical explanation of them is highly desirable.

In this work, the GMR in magnetic superlattices is calculated by using the real space two-point conductivity formula [9,10]. The present investigation is confined to the systems where the spin dependence of the scattering at the interfaces is stronger than that in the bulk of FM layers. We will focus our attention on two aspects. One is the strength effect of interfacial scattering on

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the GMR. It is found that, if the enhanced scattering asymmetry at interfaces is fixed, with increasing the strength of the interfacial scattering, the GMR amplitude first increases and then decreases, exhibiting a novel maximum. This theoretical result can be used to reconcile the superficially contradictory experimental observations mentioned above [2–4]. The other interest is the dependence of the GMR on the angle θ between the magnetizations in two successive FM layers. The GMR amplitude is found to obey an approximately linear dependence on $\cos^2\theta$ provided that the layer thicknesses are not very small. In the thin layer case, deviations from this dependence will grow larger with decreasing the layer thickness.

Let us consider a FM/NM superlattice in which the NM layers of thickness a and the FM layers of thickness b are alternately stacked along the z axis. The electrical transport with currents in the plane of the layers can be described by the two-point conductivity formula [9,10]

$$J(z) = \int_{-\infty}^{\infty} \mathrm{d}z' \sigma_{\parallel}(z,z') E(z'), \tag{1}$$

where

$$\sigma_{\parallel}(z,z') = \frac{3C_\mathrm{D}}{4}\int_0^1 \mathrm{d}u \frac{1-u^2}{u} \mathrm{Tr}\Big[\hat{S}(z,z')\hat{S}^{\dagger}(z,z')\Big]. \tag{2}$$

Here, $C_\mathrm{D} = n_\mathrm{e}e^2/2m$ with n_e as the electron density, and $\hat{S}(z,z') = \mathcal{P}_{z'\to z}\exp[-\int_{z^<}^{z^>}\mathrm{d}z''\hat{\lambda}^{-1}(z'')/2u]$, where $\hat{\lambda}(z)$ is the 2×2 mean free path matrix in spin space, $z^<$ ($z^>$) is the smaller (larger) one of z and z', and the path ordering operator $\mathcal{P}_{z'\to z}$ reorders the noncommutating 2×2 scattering matrices $\hat{\lambda}^{-1}(z'')$ from z to z' and from right to left. This conductivity formula can be derived either from the real-space Kubo formula under the quasiclassical approximation [9] or directly from the Boltzmann equation approach [10]. It is worthy to be mentioned that equation (2) is applicable not only to a collinear magnetization configuration, but also to arbitrary magnetization configurations. So, it can be used to study the magnetization dependence of the GMR. For the transport parallel to the plane of the layers, since $E(z)\equiv E$ is a constant independent of z, we can obtain from equation (1) for the average conductivity

$$\sigma = \frac{1}{L}\int_0^L \mathrm{d}z \int_{-\infty}^{\infty}\mathrm{d}z'\sigma_{\parallel}(z,z'), \tag{3}$$

where L stands for the minimum period of the spatial distribution of the current density.

We now specify the input parameters which will be used in our calculations. The mean free path matrix in equation (2) is given by [10]

$$\hat{\lambda}^{-1}(z) = \frac{1}{2}\left(\frac{1}{\lambda^{\uparrow}(z)}+\frac{1}{\lambda^{\downarrow}(z)}\right) + \frac{1}{2}\left(\frac{1}{\lambda^{\downarrow}(z)}-\frac{1}{\lambda^{\uparrow}(z)}\right)\hat{\boldsymbol{\sigma}}\cdot\mathbf{m}(z), \tag{4}$$

where $\lambda^{\uparrow}(z)$ and $\lambda^{\downarrow}(z)$ are the position-dependent mean free paths for spin-up and spin-down electrons, respectively, $\hat{\boldsymbol{\sigma}}$ is the Pauli spin vector operator and $\mathbf{m}(z)$ is a unit vector along the direction of the local magnetization. In the FM layers, we denote $\lambda^{\uparrow}(z) = \lambda^{\uparrow}_{\mathrm{FM}}$ and $\lambda^{\downarrow}(z) = \lambda^{\downarrow}_{\mathrm{FM}}$ with bulk scattering asymmetry defined as $N_{\mathrm{FM}} = \lambda^{\uparrow}_{\mathrm{FM}}/\lambda^{\downarrow}_{\mathrm{FM}}$; while in the NM layers, the scattering is spin-independent so that we have $\lambda^{\uparrow}(z) = \lambda^{\downarrow}(z) = \lambda_{\mathrm{NM}}$. Furthermore, the interfacial scattering is modeled as the bulk scattering in a thin region where the FM and NM atoms are mixed. In such a mixing layer, we denote $\lambda^{\uparrow}(z) = \lambda^{\uparrow}_{\mathrm{I}}$ and $\lambda^{\downarrow}(z) = \lambda^{\downarrow}_{\mathrm{I}}$ with interfacial scattering asymmetry $N_{\mathrm{I}} = \lambda^{\uparrow}_{\mathrm{I}}/\lambda^{\downarrow}_{\mathrm{I}}$. The thickness d of the mixing layer is assumed to be much smaller than a and b. In this case, only the ratio between d and the mean free path in the mixing layer is relevant to physical results, so that a dimensionless parameter $\gamma_{\mathrm{I}} = d/\lambda^{\uparrow}_{\mathrm{I}}$ can be introduced to characterize the strength of the interfacial scattering. As a result, the independent input parameters for the problem are

(1) a, λ_{NM},	for NM layers;
(2) b, $\lambda^{\uparrow}_{\mathrm{FM}}$, N_{FM},	for FM layers;
(3) γ_{I}, N_{I},	for interfaces.

Since we have assumed that the mean free paths are z independent within each layers, the integral over z and z' in equation (3) is readily performed and the average conductivity is obtained as

$$\sigma = \frac{3C_D}{2L}\int_0^1 \mathrm{d}u(1-u^2)\sum_{n=1,2\cdots}\mathrm{Tr}\Big[d_n\hat{\lambda}_n - u(1-\hat{E}_n)\hat{\lambda}_n^2 + 2u(1-\hat{E}_n)\hat{\lambda}_n\sum_{m(>n)}\hat{S}_{nm}(1-\hat{E}_m)\hat{\lambda}_m\hat{S}^{\dagger}_{nm}\Big], \tag{5}$$

where d_n and $\hat{\lambda}_n$ represent the thickness and mean free path matrix in the nth layer, respectively, $\hat{E}_n = \exp(-d_n\hat{\lambda}_n^{-1}/u)$, and $\hat{S}_{nm} = \hat{S}(z,z')$ with z (z') at the right (left) surface of the nth (mth) layer. The summation over n in equation (5) is confined in a single period L. The matrix multiplication will be numerically evaluated.

Role of interfacial scattering

In Figure 1, the maximal GMR ratio $\Delta\sigma/\sigma_{\mathrm{FM}}$ is plotted as a function of the interfacial scattering strength γ_{I} for different values of layer thicknesses and interfacial scattering asymmetry N_{I}. Here, $\Delta\sigma = \sigma_{\mathrm{FM}} - \sigma_{\mathrm{AF}}$, where σ_{FM} and σ_{AF} are the spatial averages of the conductivity in the FM and antiferromagnetic configurations, respectively. It is found that increased spin dependence of interfacial scattering always leads to an enhancement of the GMR ratio. The GMR increase is relatively larger in the multilayer with thinner layers, where the interfacial scattering occupies a greater weight in producing the GMR. What is more important, we find that all the $\Delta\sigma/\sigma_{\mathrm{FM}}$ *vs.* γ_{I} curves, with

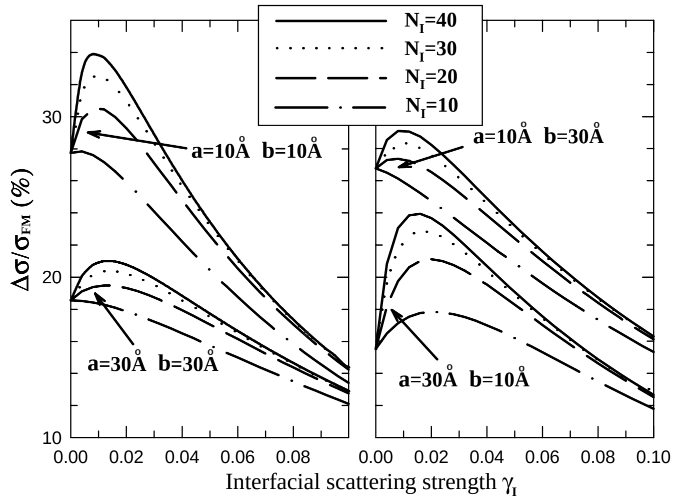

Fig. 1. Maximal GMR ratio $\Delta\sigma/\sigma_{\rm FM}$ as a function of interfacial scattering strength $\gamma_{\rm I}$ for several values of layer thicknesses and interfacial scattering asymmetry as labeled in the figure. The other parameters used are $N_{\rm FM} = 10$ and $\lambda_{\rm NM} = \lambda_{\rm FM}^{\uparrow} = 200$ Å.

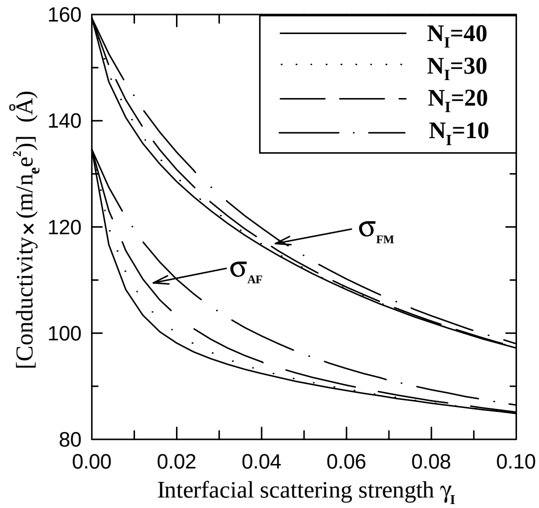

Fig. 2. Conductivity for ferromagnetic configuration $\sigma_{\rm FM}$ and antiferromagnetic configuration $\sigma_{\rm AF}$ as a function of interfacial scattering strength $\gamma_{\rm I}$. Here, $a = 30$A, $b = 10$A and other parameters are taken as the same as in Figure 1.

enhanced interfacial scattering asymmetry ($N_{\rm I} > N_{\rm FM}$), exhibit novel peak behavior. As $\gamma_{\rm I}$ is increased, the GMR first increases and then decreases, strongly suggesting that there exists an optimum value of $\gamma_{\rm I}$ at which there is a maximal GMR amplitude. For the $a = 30$ Å and $b = 10$ Å sample, we also show the conductivity values $\sigma_{\rm FM}$ and $\sigma_{\rm AF}$ in Figure 2. It is interesting to notice that both $\sigma_{\rm FM}$ and $\sigma_{\rm AF}$ change monotonously with changing the interfacial scattering strength. Therefore, the GMR peak discussed above originates from the nonmonotonous variation of $\Delta\sigma$. In the present calculation, the spin asymmetric factor $N_{\rm FM}$ is taken to be $N_{\rm FM} = 10$, which is close to the value 11.8 obtained in the quantum GMR theory [7] by fitting the experimental data for Fe/Cr superlattices. For other systems the parameter may be different. For example for Co/Ag multilayers, Pratt *et al.* and Schroeder *et al.* [11] found $N_{\rm FM} \simeq 3$ based on a resistor network analysis within a two fluids model, being much smaller than that for the Fe/Cr superlattices. Nevertheless, our results in particular the peak behavior of the GMR will not change qualitatively with changing the values of the input parameters, as long as the enhanced spin asymmetry of the interfacial scattering is taken into account.

The peak behavior of the GMR can be understood as a result of the competition between the following two factors. On the one hand, an enhancement of interfacial scattering strength will increase the proportion of the interfacial scattering to the total scattering. Since the scattering asymmetry at the interface is greater than that in the bulk of the layers, enhanced interfacial scattering should increase the GMR effect. On the other hand, as the interfacial scattering is increased strong enough, its further increase may prevent conduction carriers from moving from one FM layer to the others and so the electronic transport parallel to the plane of the layers become insensitive to the change in the magnetization configuration of the FM layers, which is unfavorable to the GMR effect. In the limiting case where the interfacial scattering is completely diffusive for both spin channels, the interfaces can be regarded as thin isolator films, and each layer contributes to the conductivity independently so that the GMR effect vanishes. Such a competition between the two opposite effects can produce the peak behavior of the GMR ratio. It is interesting to see that for the set of curves with the smallest ratio b/a ($a = 30$ Å and $b = 10$ Å) in Figure 1, the peak behavior persists even for the case $N_{\rm I} = N_{\rm FM} = 10$. This is because with decreasing b/a, the spin-dependent proportion of the bulk scattering decreases and so the spin-dependent interfacial scattering plays more important role in the GMR effect.

The present theoretical result can account qualitatively for the inconsistent experimental data [2–4] by linking $\gamma_{\rm I}$ with interfacial roughness. Taking into account that there exists an optimal value of interfacial roughness, we may argue that the experimental result that increased interfacial roughness enhances the GMR [2] is obtained in the multilayers whose interfacial roughness is below its optimal value, while the opposite experimental result [3,4] may be observed for the interfacial roughness above the optimum value. Besides, the interfacial roughness may affect the interfacial scattering asymmetry, this effect depending mainly on what types of atoms are mixed at the interfaces. As an example, for the Fe/Cr multilayer increased mixing of Fe and Cr atoms at the Fe/Cr interfaces will increase both the interfacial scattering strength and spin asymmetry, which is another reason for that its GMR can be enhanced by increased interfacial roughness [2].

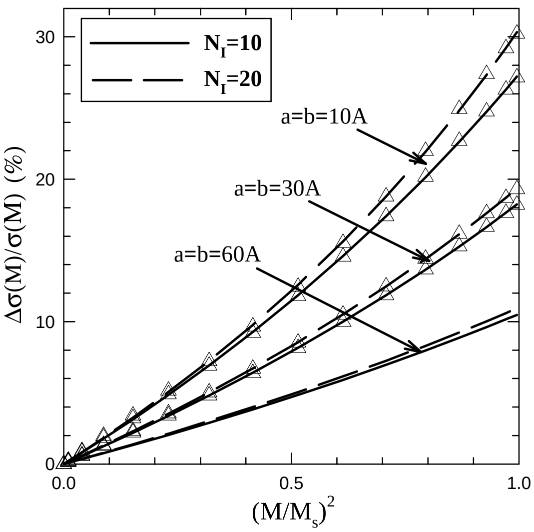

Fig. 3. GMR amplitude $\Delta\sigma(M)/\sigma(M)$ as a function of $(M/M_{\rm s})^2$. The interfacial scattering strength is taken to be $\gamma_{\rm I} = 0.01$. The other parameters are $N_{\rm FM} = 10$ and $\lambda_{\rm NM} = \lambda^{\uparrow}_{\rm FM} = 200$A. Triangles represent the best fits of equation (6) to the magnetization-dependent GMR calculated from equation (5).

Angle dependence of GMR

For simplicity, we consider that a magnetic multilayer has the canted magnetization configuration in the presence of an external magnetic field that is smaller than the saturation one, in which the magnetization direction of the nth FM layer may be characterized as the polar angles $[\theta, \phi_n]$ with $\phi_n = 0$ and π for n being odd and even, respectively. It is easy to see that 2θ corresponds to the angle between the magnetizations in two successive magnetic layers. For this particular magnetic configuration, the global magnetization M of the system is given by $M = M_{\rm s}\cos\theta$, where $M_{\rm s}$ is the saturation magnetization of the system in the presence of a saturation magnetic field. Therefore the angle dependence is actually equivalent to a magnetization dependence. In general, the magnetization dependent GMR can be defined as $\Delta\sigma(M)/\sigma(M)$ with $\Delta\sigma(M) = \sigma(M) - \sigma(0)$. In Figure 3, we plot the calculated GMR ratio as a function of $(M/M_{\rm s})^2 = \cos^2\theta$ for different values of layer thicknesses and interfacial scattering asymmetry. For the system with relatively thick layers ($a = b = 60$ Å), the GMR ratio obeys an approximate linear dependence on $(M/M_{\rm s})^2$. With decreasing the layer thickness, small departures from this dependence can be seen. In this case, the calculated GMR amplitude $\Delta\sigma(M)/\sigma(M)$ is found to be a relatively complicated even function of $M/M_{\rm s}$, which can be expressed as

$$\frac{\Delta\sigma(M)}{\sigma(M)} = C\left[\left(\frac{M}{M_{\rm s}}\right)^2 + \alpha\left(\frac{M}{M_{\rm s}}\right)^4\right]. \qquad (6)$$

With adjusted parameters C and $\alpha\,(< 0.5)$, its best fits to the calculated results (solid and dashed lines) are given by triangles in Figure 3.

The magnetization dependence of the GMR ratio has been studied experimentally in several magnetic layered structures [12–14], and it has been found that the GMR ratio varies approximately as $(M/M_{\rm s})^2$. On the theoretical side, Vedyayev *et al.* [15] and Barnaś *et al.* [16] have studied angular dependence of the GMR in FM bilayers and FM/NM/FM sandwiches. They showed the linear dependence of GMR on $(M/M_{\rm s})^2$ when there is no potential step at interfaces. They also suggested that in the presence of potential steps at the interfaces, significant deviations from the linear behavior can be found. For magnetic superlattices, we have previously reproduced this linear dependence analytically from the Boltzmann equation approach in two limiting cases [10]. One is the thick layer limit, where the thickness of the FM layer is larger than the mean free paths $b > \lambda^{\rm s}_{\rm FM}$, the other is the homogeneous limit of $d_n \ll \lambda^{\rm s}_n$. The latter limit may not be easily realized experimentally, for there exists relatively strong interfacial scattering in real systems. The present numerical result shows that, between the two limiting cases, there are obvious deviations from the linear dependence on $(M/M_{\rm s})^2$. Such deviations from the linear behavior in magnetic superlattices seem intrinsic, since their presence does not depend on whether the potential steps at interfaces are taken into account. They occur whenever the electrons can propagate through three or more magnetic layers within an average mean free path. The magnetization dependence of the GMR given in equation (6) has been observed in magnetic granular systems [17]. It is expected that the predicted deviations from the linear dependence on $(M/M_{\rm s})^2$ can be observed in future experiments of magnetic superlattices composed of sufficiently thin layers.

In summary, we have numerically studied the effects of the interfacial roughness on the GMR and the magnetization dependence of the GMR. The enhanced scattering asymmetry at the interfaces as predicted in reference [8] has been taken into account in the present theory. It is found that there exists an optimum degree of interfacial roughness at which the GMR ratio exhibits its maximum. This theoretical result can be used to settle a dispute in experimental reports. In magnetic superlattices with relatively thick layers, the GMR amplitude obeys an approximate linear dependence on $(M/M_{\rm s})^2$, but obvious deviations from this linear dependence appear in systems with sufficiently thin layers.

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