Enhancement of GMR due to spin-mixing in magnetic multilayers with a superconducting contact

F. Taddei*,
School of Physics and Chemistry, Lancaster University, Lancaster, LA1 4YB, UK

S. Sanvito†,
Materials Department, University of California, Santa Barbara, CA 93106, USA

C.J. Lambert‡
School of Physics and Chemistry, Lancaster University, Lancaster, LA1 4YB, UK

(Received 1 November 2018)

We study the Giant Magnetoresistance (GMR) ratio in magnetic multilayers with a single superconducting contact in the presence of spin-mixing processes. It has been recently shown [1] that the GMR ratio of magnetic multilayers is strongly suppressed by the presence of a superconducting contact when spin-flipping is not allowed. In this Letter we demonstrate that the GMR ratio can be dramatically enhanced by spin-orbit interaction and/or non-collinear magnetic moments. The system is described using a tight-binding model with either s-p-d or s-d atomic orbitals per site.

PACS numbers: 75.70.Pa, 74.80.Dm

Hybrid nanostructures form a fascinating melting pot for studying the interplay between fundamental quantum phenomena, often revealing new and unexpected physics. One recently-recognized class of such structures, involving the coexistence of superconducting contacts and ferromagnetic domains, has led to the identification of a number of fundamental issues [2–8], several of which are currently unresolved. In this Letter, we examine one such issue, posed by experiments on giant magnetoresistance (GMR) in magnetic multilayers with superconducting (S) contacts and current-perpendicular-to-the-plane (CPP). Recognizing that the sub-gap conductance of such structures is mediated by Andreev scattering, it was recently noted [1] that in the absence of spin-flip processes, the conductance of a metallic (i.e. diffusive) multilayer in the presence of aligned magnetic moments is almost identical to that of the multilayer when adjacent moments are anti-aligned and therefore the conventional GMR ratio should be strongly suppressed. Since large GMR ratios are observed experimentally [9], it is clear that even a qualitative understanding of transport in such structures must incorporate the effects of spin-mixing. The aim of this Letter is to present the first theoretical description of CPP GMR in M-multilayers mixing. The system under consideration is a disordered magnetic multilayer consisting of an alternating sequence of magnetic layers each of length $l_M$ and non-magnetic layers (N) of length $l_N$. The building block of the magnetic structure is the bilayer $[M/N]$ of length $l_B = l_N + l_M$. The magnetic moments of even-numbered M-layers make an angle $\theta$ relative to those of odd-numbered M-layers. Experimentally, $\theta$ can be varied by applying an external magnetic field with antiparallel (AP) alignment ($\theta = \pi$) typically occurring at zero field and parallel (P) alignment ($\theta = 0$) at large enough fields. The current flows perpendicular to the planes of the multilayer, which makes contact with a metallic normal lead on the left-hand side of the multilayer and a superconducting lead on the right-hand side. GMR is the drastic increase in electrical conductance $G(\theta)$ that occurs when the system switches from the AP to the P alignment with the conventional GMR ratio defined by: $\rho = \frac{G(0) - G(\pi)}{G(0)}$.

Following [1], the multilayer and leads are modelled using a tight-binding Hamiltonian on a cubic lattice with on-site energies a random number in the range $[-\frac{\Delta}{2}, +\frac{\Delta}{2}]$. The on-site Hamiltonian has the following structure:

$$H = \begin{pmatrix}
H_{\uparrow}^\dagger & \mu_{xy} & \Delta & 0 \\
\mu_{xy}^\ast & H_{\downarrow}^\dagger & 0 & -\Delta \\
\Delta^\dagger & 0 & H_{\downarrow}^\dagger & -\mu_{xy}^\ast \\
0 & -\Delta^\dagger & -\mu_{xy} & H_{\uparrow}^\dagger
\end{pmatrix}$$

(1)

where $H_{\uparrow(\downarrow)}^\dagger$ is the Hamiltonian for up (down)-spin particles ($s$ and $d$ bands), $H_{\downarrow(\uparrow)}^\dagger = -H_{\uparrow(\downarrow)}^\dagger$* is the Hamiltonian for up(down)-spin holes and $\Delta$ is the superconducting order parameter. Here $\mu_{xy} = -\mu_x + i\mu_y$, where

*e-mail: f.taddei@lancaster.ac.uk
†e-mail: ssanvito@mrl.ucsb.edu
‡e-mail: c.lambert@lancaster.ac.uk
\( \mu_{x(y)} \) is the \( x(y) \)-component of the exchange field \( \vec{\mu} \). Note that \( \vec{\mu} \) is non-zero only for electrons in the \( d \)-band in the M-layers and \( \Delta \) is non-zero only in the right-hand-side superconducting lead. Within the tight-binding formulation, SO interaction can be included by adding to the Hamiltonian the following term:

\[
V_{SO} = V_1 \sum_{i,j,\alpha,s} \vec{\sigma} \cdot \vec{R}_{i,j} c_{\alpha,i}^{\sigma} c_{\alpha,j}^{-\sigma} \tag{2}
\]

where \( V_1 \) is a constant which determines the interaction strength, \( \vec{\sigma} \) is a vector of Pauli matrices, \( \vec{R}_{i,j} \) is the unit vector which connects site \( i \) with the neighbouring site \( j \). \( c_{\alpha,i}^{\sigma} \) is the annihilation operator for electrons of spin \( \sigma \) in the \( \alpha \) \( (s, d) \)-band on site \( i \). In the presence of disorder \( \rho \), one produces spin-flip scattering since it couples electrons with different spin on neighbouring sites.

In the presence of disorder, to study the largest possible sample cross sections, we consider 2 orbitals per site, which is the minimal model capable of reproducing scattering potential at the N/M interface and interband scattering \( \theta \). The tight-binding parameters are chosen to reproduce the GMR ratio and conductances obtained from an ab initio material specific calculation for Cu/Co multilayers \( \theta \).

In the presence of spin-flip scattering the two-spin-fluid approximation does not hold and the zero-temperature, zero-bias, normal-state Landauer formula takes the form:

\[
G^{NN} = \frac{e^2}{h} \sum_{\sigma} \text{Tr} \left\{ t_{\sigma\sigma'} t_{\sigma\sigma'} \right\} \tag{3}
\]

where \( t_{\sigma\sigma'} \) is the matrix of transmission amplitudes for injected \( \sigma' \)-spin electrons in the left-hand lead into \( \sigma \)-spin electrons in the right-hand lead. When the right-hand lead is in the superconducting state, the conductance is given by \( \theta \):

\[
G^{NS} = \frac{e^2}{h} \sum_{\sigma} \text{Tr} \left\{ r_{\sigma\sigma'} r_{\sigma\sigma'} \right\} \tag{4}
\]

where \( r_{\sigma\sigma'} \) is the Andreev reflection matrix for injected \( \sigma' \)-spin electrons in the left-hand lead to be reflected into \( \sigma \)-spin holes. In what follows, the scattering amplitudes are calculated exactly by solving the Bogoliubov-de Gennes equation using an efficient recursive Green’s function technique \( \theta \).

We shall now turn to the central results of this Letter, namely that in the presence of strong-enough spin-mixing, either produced by SO coupling or non-collinear moments, GMR in the presence of a S-contact approaches that of two normal contacts, with values of \( \rho \) of the order of 100 \%. First consider the effect of SO coupling within the multilayer. Fig. 2 shows the conventional GMR ratio as a function of the SO interaction strength \( V_1 \) for a disordered multilayer of 40 bilayers, with \( l_M = 15 \) and \( l_N = 8 \).

As expected, in the NN case \( \rho \) decreases monotonically from \( \sim 200 \% \) at \( V_1 = 0 \), to zero at large \( V_1 \) (\( \sim 0.17 \) eV). In contrast for the NS case, \( \rho \) initially increases with increasing \( V_1 \), eventually joining the NN curve at \( V_1 \sim 0.08 \) eV. As a second source of spin-mixing, consider the effect of non-collinear magnetic moments when \( V_1 = 0 \). Fig. 3 shows the \( \theta \)-dependence of the GMR ratio defined as \( \rho(\theta) = \frac{G(\theta) - G(\pi)}{G(\pi)} \). Whereas in the NN case \( \rho(\theta) \) decreases monotonically with increasing \( \theta \), in the NS case \( \rho(\theta) \) exhibits a pronounced maximum around \( \theta = \pi/8 \).

To understand these results, first consider the case of non-collinear moments. In the presence of two normal-metallic contacts, the conductance \( G(\theta) \) has been theoretically studied in \( \| \) where it is predicted that \( G(\theta) - G(\pi) \) tends monotonically to zero as \( \theta \) varies from 0 to \( \pi \). In addition, the dependence of the resistance on the angle \( \theta \) has been experimentally found \( \| \) to contain a term proportional to \( \cos^2(\theta/2) \) and a second term proportional to \( \cos^4(\theta/2) \). In the presence of a S-contact, where \( G(0) \approx G(\pi) \), this behaviour is drastically changed by the presence of an extremum which occurs at some intermediate angle \( \theta_c \), the value of which depends on the interplay between competing effects. Since \( \theta(H) \) is a function of the applied magnetic field \( H \), the presence of a S-contact introduces a new characteristic field \( H_c \) for which \( \theta(H_c) = \theta_c \). For a disordered multilayer of 22 bilayers, with \( l_M = 30 \), \( l_N = 16 \), the insert in Fig. 2 shows the \( \theta \)-dependence of the conductance divided by the number of open channels in the normal lead. As expected \( G^{NN}(\theta) \) is a monotonic function of \( \theta \), whereas \( G^{NS}(\theta) \) possesses an extremum at \( \theta_c \approx \pi/8 \). To understand why the extremum is a maximum, recall that for \( \theta = 0 \) or \( \theta = \pi \), when spin is conserved, current flows when a right-going (spin \( \sigma \)) electron passes through the multilayer, Andreev reflects as a left-going (spin \( -\sigma \)) hole, which retraverses the multilayer. A M-layer whose moment is aligned with the spin of the incident electron is anti-aligned with the spin of the outgoing hole and consequently the number of aligned and anti-aligned M-layers encountered by a given quasi-particle is the same for both \( \theta = 0 \) and \( \theta = \pi \) (only the order differs). When the elastic mean free path is comparable with the total multilayer length, the resistance of traversed layers add in series and therefore, apart from small differences due to interference effects \( \| \), \( G^{NS}(0) \approx G^{NS}(\pi) \). Furthermore, since a quasi-particle must necessarily traverse regions in which it is a minority spin, both \( G^{NS}(0) \) and \( G^{NS}(\pi) \) are low-conductance states. In contrast, as \( \theta \) increases from zero, this conductance bottleneck is removed, because an Andreev reflected minority hole can spin-convert to a majority hole, thereby avoiding anti-aligned moments on its return journey. Of course this initial increase in \( G^{NS}(\theta) \) is eventually overcome by the usual GMR effect which decreases \( G^{NS}(\theta) \) as \( \theta \to \pi \), thereby producing an overall maximum.
In the absence of disorder, the nature of the extremum is determined by interface scattering and band structure. To illustrate this consider a clean multilayer which is perfectly periodic and therefore the variation of the conductance with \( \theta \) arises from tuning of the ballistic spin-filtering by the structure. Fig. 1 shows the conductance divided by the number of open channels in the left-hand-side normal lead. As expected, \( G^{NN}(\theta) \) is a monotonous function of \( \theta \), whereas \( G^{NS}(\theta) \) exhibits a minimum around \( \pi/2 \) and then increases. (In this case, translational invariance in the transverse direction allowed us to use a full \textit{ab initio}, \textit{spd} Hamiltonian to obtain the results of Fig. 1.) In the NN case, the dependence of the multilayer resistance on \( \theta \) predicted by our model is in good agreement with experiment \cite{16}. In Ref. \cite{16} the ratio between the resistance at a given \( \theta \) and the resistance with AP alignment has been found to fit the following function:
\[
\frac{R(\theta)}{R(\pi)} = 1 - a \cos^2(\theta/2) + b \cos^4(\theta/2)
\]
where \( a \) and \( b \) are fitting constants. In Fig. 1 we show the plot of such a ratio for the disordered multilayer considered above, along with the best fit to function (5). In addition we also checked that this ratio cannot be fitted with the same accuracy assuming a pure dependence on \( \cos^2(\theta/2) \) (i.e. with \( b = 0 \)). For \( G^{NS}(\theta) \) however, no such analytic results currently exist.

Let us now turn attention to the effect of SO coupling. Figs. 2a and 2b show the conductances as a function of the SO strength \( V_1 \) for, respectively, the NN and the NS case. In the NN case (Fig. 2a) \( G^{NN}_P \) decreases as \( V_1 \) increases and eventually joins the curve for \( G^{NN}_A \). This can be understood in terms of the heuristic model presented in Ref. 4, because, as the SO strength increases, the average length required for a spin to flip (spin relaxation length \( \lambda_{sf} \)) gets shorter. Therefore in the P alignment, an injected majority electron travels through the multilayer for a length \( \lambda_{sf} \) before being scattered into a minority spin, thereby producing a decrease in the conductance. This suggests that the value of \( V_1 \) for which \( G^{NN}_P \approx G^{NN}_A \) corresponds to a spin relaxation length \( \lambda_{sf} \) close to the period \( l_B \) of the multilayer. We have carried out a range of simulations which show that this value of \( V_1 \) does not depend on the overall length of the multilayer, but decreases with increasing \( l_B \). As expected, the conductance with AP alignment does not change significantly with \( V_1 \).

In the NS case (Fig. 2b) the conductance in the P aligned state rapidly increases with \( V_1 \), reaching a maximum and thereafter decreases, eventually joining the curve for the AP configuration. Clearly the enhancement in \( G^{NS}_P \) is produced by the onset of spin-flip scattering. The abrupt increase is understandable, since even a relatively small probability for spin flipping opens a highly conductive “channel” if the spin-flip events take place in the vicinity of the interface. As one can see in the insert of Fig. 2 for larger values of \( V_1 \), the conductance \( G^{NS}_P \) joins \( G^{NN}_P \) and together they decrease thereafter. As in the normal case \( G^{NS}_{AP} \) depends weakly on \( V_1 \). The value \( V_1 \approx 0.08 \), at which \( G^{NS} \) is maximum, corresponds to a spin relaxation length close to the total length of the multilayer and, as expected, separate simulations show that this value of \( V_1 \) decreases with increasing total length. Similarly the value of \( V_1 \) at which the GMR ratio (of Fig. 1) vanishes corresponds to a spin-relaxation length of the order the bilayer thickness \( l_B \) and is independent of the total length of the system.

In conclusion, we have demonstrated that spin-mixing plays a crucial rôle in determining both the qualitative and quantitative features of GMR in magnetic multilayers with a S-contact. In contrast with the normal case, where spin-mixing suppresses GMR, we find that the GMR ratio can be dramatically enhanced by the presence of spin-orbit interactions and/or non-collinear magnetic moments. In experiments carried out to-date, the presence of large spin-orbit scattering \cite{18} presumably masks the mechanism shown in Fig. 1 which is predicted to be a generic feature in the absence of other spin-mixing processes. This suggests that lighter metals and superconductors would be more appropriate for observing the new extrema predicted in this Letter. Finally we note that for the future it would be of interest to examine spin-mixing in non-diffusive NS structures such as clean spin-valves \cite{19}, where the GMR ratio can be non-zero or negative, even in the absence of spin-flip processes.

**FIG. 1.** Conventional GMR ratio as a function of the SO interaction strength for NN and NS cases. Results correspond to disordered multilayers (\( W = 0.6 \text{ eV} \)) comprising 40 bilayers with \( l_M = 15 \) and \( l_N = 8 \). Samples are formed by repeating \((3 \times 3)\) disordered unit cells in the transverse plane and summing over 25 k-points in the 2-dimensional Brillouin zone. The points are an average over 20 disorder realizations and the error bars represent the standard deviations from the mean. In the insert, comparison between the conductances in the P alignment for the NN and NS cases as functions of the SO interaction strength.
Fig. 2. $\theta$-dependent GMR ratio for NN and NS cases in the absence of SO coupling. Results correspond to disordered multilayers ($W = 0.6$ eV) of 22 bilayers with $l_M = 30$, $l_N = 16$, considering a $(3 \times 3)$ unit cell in the transverse plane, and a sum over 25 k-points in the 2-dimensional Brillouin zone. The points are the average over 50 realizations of disorder. In the insert, $\theta$-dependent conductance for NN and NS cases in the absence of SO coupling.

Fig. 3. $\theta$-dependent conductance for NN and NS cases in the absence of SO coupling for a clean multilayer. The multilayer is modelled by a material-specific spd-band Hamiltonian (see Ref. [1]), with Co as M-material, Cu as N-material and Pb as S-material. $l_M = 7$ and $l_N = 10$, considering a $(1 \times 1)$ unit cell in the transverse plane, summing about 5000 k-points in the 2-dimensional Brillouin zone.

Fig. 4. Plot of the ratio $\frac{R(\theta)}{R_{\pi}}$ along with the best fit to the function $f$. The value of the fitting parameters are: $a = 0.407$, $b = -0.238$. Results correspond to a disordered multilayer with the same parameters as in Fig. 2.

Fig. 5. Conductances for P and AP alignments as functions of the SO interaction strength for the NN case (a) and for the NS case (b). Results correspond to a disordered multilayer with the same parameters as in Fig. 1.

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