Backflow Effect on Spin Diffusion Near Ferromagnet-Superconductor Interface

M. Faiz, R.P. Panguluri and B. Nadgorny  
*Department of Physics and Astronomy, Wayne State University, Detroit, Michigan 48201*

B. Balke  
*Institute for Materials Science, University of Stuttgart, 70569 Stuttgart, Germany*

S. Wurmehl  
*Institute for Materials Research IFW, 01069 Dresden, Germany*

C. Felser  
*Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*

A. G. Petukhov  
*Google Inc., Venice, California 90291, USA*  
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The behavior of spin propagation in metals in various measurement schemes is shown to be qualitatively different than a simple exponential decay - due to the backflow effect on spin diffusion in the presence of interfaces. To probe this effect we utilize the spin sensitivity of an Andreev contact between gold films of variable thickness deposited on top of a spin injector, Co$_2$Mn$_{0.5}$Fe$_{0.5}$Si, with the spin polarization of approximately 45%, and Nb superconducting tip. While the results are consistent with gradually decaying spin polarization as the film thickness increases, the spin diffusion length in Au found to be 285 nm, is more than two times larger that one would have obtained without taking the backflow effect into account.

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Processes of spin injection and spin accumulation are of fundamental importance for operation and underlying physics of spintronic devices [1]. After it was realized that a spin polarized current can induce non-equilibrium spin populations of both nuclear [2] and electronic [3] subsystems in a normal (non-magnetic) metal, the related problem of spin injection from a ferromagnet (F) into a normal metal (N) was considered by Aronov [4]. Johnson and Silsbee [5] performed the first measurements of spin relaxation in a purely electronic subsystem. These experiments utilized the so-called lateral non-local geometry to determine a spin diffusion length in aluminum by probing a difference between chemical potentials of the two spin subbands. A more convenient version of this technique was later adopted for F/N/F structures [6], and has been further developed by Jedema et al. [7]. Another means to determine spin-diffusion length in metals is to analyze the thickness dependence of the current-perpendicular-to-plane (CPP) giant magnetoresistance (GMR) effect [8, 9]. Finally, an optical technique based on measuring the spin accumulation via the Kerr effect has been successfully implemented by Crooker et al. [10].

Most of the measurement techniques described above use the implicit assumption that spin in a normal metal decays exponentially with distance. While in the case of spin injection into a normal metal of infinite thickness this assumption is correct, the presence of a spin selective interface within a distance that is comparable to the spin diffusion length would modify this dependence in any real measurements. Indeed, a spin selective interface imposes different boundary conditions for spin-up and spin-down electrons, thus resulting in a backflow of spin polarized electrons away from that interface.

The backflow effect exists in the case of an N/F interface and thus have significant implications for the description of spin accumulation and spin propagation in GMR devices, but it arguably can be the most pronounced in the case of N/S interface. At the energies below the superconducting gap $\Delta$ and temperatures far enough from the superconducting transition temperature $T_c$, Andreev reflection [11] is the dominant process [12] that allows quasiparticle current propagation from a normal metal into a superconductor by converting quasiparticles with opposite spins into Cooper pairs. Any asymmetry in the quasiparticle spin balance, that may exist, for example in a ferromagnet, would reduce the probability of such a process and consequently the conductance across the interface [13]. Based on this property of Andreev reflection at an F/S interface it has been shown that the junction conductance is sensitive to the values of spin polarization in a ferromagnet [14, 15]. Similarly, a spin current injected into a normal metal should be sensitive to the same Andreev reflection mechanism due the non-equilibrium spin accumulation near an N/S interface. Such spin accumulation will gradually decrease as we increase the thickness of the N-layer [16].

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In this Letter we propose to use Point Contact Andreev Reflection (PCAR) spectroscopy to investigate the backflow effect on spin diffusion and spin accumulation by exploiting the dependence of the magnitude of this effect on metal thickness, as shown in Fig. 1. In particular, we use spin injection from a highly spin polarized Heusler alloy, Co$_2$Mn$_{0.5}$Fe$_{0.5}$Si into gold films of different thicknesses to observe a gradual decay of spin polarization in Au. We formulate a phenomenological description of such transport in a diffusive regime to determine the spin diffusion length $L_N$ in gold and demonstrate that a combination of the PCAR technique with the proper phenomenological theory could result in an alternative electrical technique for probing spin diffusion length in normal metals.

As most of the Heusler alloys [17] Co$_2$Mn$_{0.5}$Fe$_{0.5}$Si has a high (~1000K) Curie temperature and is believed to be fairly highly spin polarized. The samples of the Heusler alloy Co$_2$Mn$_{0.5}$Fe$_{0.5}$Si were fabricated by arc melting from stoichiometric ratio of constituents in an argon atmosphere of $10^{-4}$ mbar. After subsequent annealing of the polycrystalline ingots in an evacuated quartz tube at 1273K for 21 days the samples with the Heusler type L21 structure were obtained, as was verified by X-ray powder diffraction (XRD) using Mo K$_\alpha$ excitation. Flat disks were then cut from the ingots and polished before removing the native oxide by Ar$^+$ ion bombardment. The sample composition was further verified by X-ray photoemission (ESCA) with no impurities detected. Gold films of 99.99% purity and variable thicknesses (from 7 nm to 475nm) were then deposited on the polished surface of the disks by thermal evaporation in vacuum, immediately followed by the PCAR measurements.

The measurements of the structure shown in Fig. 1 were performed in the point contact geometry with Nb superconducting tips. The tips were fabricated by the standard electrochemical etching of 250 $\mu$m Nb wire, as described in Ref. [18]. Using freshly etched Nb tips and oxide-free Au film helped to facilitate the establishment of a stable contact (on the order of 50-100 $\Omega$), typically without the need of further adjustments, thus largely alleviating any concerns of tip-film mechanical interference; additionally post-measurement microscopy of the contact area was performed. The current–voltage ($I-V$) and the differential conductance $dI/dV$ measurements were performed by a standard four-probe technique as described in detail in Ref. [19] in the temperature range of 1.2–4.2 K. The $dI/dV$ curves are analyzed with the appropriately modified [20] Blonder-Tinkham-Klapwijk (BTK) weak coupling theory [21], with two fitting parameters, the value of spin polarization, $P$ and the interface scattering strength $Z$. First, we determined the spin polarization for bare Co$_2$Mn$_{0.5}$Fe$_{0.5}$ as an average over 15 different junctions; $P$ was found to be approximately 44 ± 3%, somewhat lower than for Co$_2$Si alloy described in earlier work [18]. For gold films deposited onto Co$_2$Mn$_{0.5}$Fe$_{0.5}$ at least ten different junctions were analyzed for each film thickness. In most cases either no or a weak $P(Z)$ dependence was observed, in the latter case $P$ was extrapolated the low $Z$ limit. In Fig. 2 four characteristic conductance curves for progressively thicker Au films are shown; the results are consistent with the notion of spin polarized current gradually decaying as the Au film thickness increases.

![FIG. 1: (Color online.) Schematics of the PCAR experiment presented in this work (left) and the next generation PCAR experiment (right)](image)

Most of the experiments on the spin injection into metals or semiconductors rely on a diffusive description of the spin transport. This is based on the fact that the spin diffusion length $L_N$ in a particular sample is related to the value of the elastic mean free path $l$ as $L_N = \frac{l}{\sqrt{\tau_s/\tau_p}}$, where $\tau_s$ and $\tau_p$ are the spin and momentum relaxation times respectively. It is generally assumed that $\tau_s \gg \tau_p$, which, in turn, justifies a description of the spin relaxation process within the diffusive transport limit. Indeed, in most metals the spin diffusion length was found to be...
roughly on the order of several hundred nanometers at low temperatures \(^9\), which is definitely larger than the typical values of the elastic mean free path.

As no spin current can propagate below the gap inside the superconductor due to the fact that only Cooper pairs with \(S = 0\) can be present there \(^{10}\), we will assume that the spin current goes to zero at the N/S interface, neglecting any possible proximity effects. In addition, we will use a 1D model to describe the spin current through the system. The validity of these assumption and their possible effect on our results will be discussed later. Our main conjecture is that the spin polarization \(P(w)\) measured in the Andreev reflection experiments is proportional to the splitting of the electrochemical potentials at the normal metal - superconductor (N-S) interface \(\Delta \zeta_N(w)\).

The splitting \(\Delta \zeta_N(x)\) is a solution of a diffusion equation: 
\[ \Delta \zeta_N(x) = A_w \exp(-x/L_N) + B_w \exp(x/L_N), \]
where \(L_N\) is the spin diffusion length of a normal metal and the coefficients \(A_w, B_w\) must be determined from the boundary conditions. The spin polarization of the current density can be expressed through \(\Delta \zeta_N(w)\) as:
\[ \gamma(x) = \frac{j^+ - j^-}{j} = \frac{\sigma_N}{2j} \frac{d\Delta \zeta_N(x)}{dx}, \]
where \(\sigma_N\) is the bulk conductivity of the normal metal. Using the boundary condition at N/S interface \(\gamma(w) = 0\) we obtain
\[ \Delta \zeta_N(w) = \frac{-\Delta \zeta_N(0)}{\cosh(w/L_N)}, \]
where \(\Delta \zeta(0)\) is the splitting of the electrochemical potentials at \(F/N\) interface. We note that \(\Delta \zeta_N(0)\) depends on \(w\) due to the positive feedback exponent. To find \(\Delta \zeta_N(0)\) we will use Rashba’s boundary condition \(^{22}\):
\[ \Delta \zeta_N(0) - \Delta \zeta_F(0) = 2jr_c [\gamma(0) - \gamma_c] \]  

Here \(\Delta \zeta_F(x)\) is the splitting of the electrochemical potentials in the ferromagnet, \(r_c = (\Sigma^+ + \Sigma^-)/(4\Sigma^+\Sigma^-)\), \(\gamma_c = (\Sigma_1 - \Sigma_\downarrow)/(\Sigma_\uparrow + \Sigma_\downarrow)\), and \(\Sigma_\uparrow, \Sigma_\downarrow\) are the contact conductances. Another boundary condition is the continuity of the spin current across \(F/N\) interface \(^{22}\):
\[ \sigma_N \Delta \zeta_N(0) - 4(\sigma_\uparrow\Sigma_\downarrow/\sigma_F) \Delta \zeta_F(0) = 2\gamma_F j, \]
where \(\gamma_F = (\sigma_\uparrow - \sigma_\downarrow)/\sigma_F\), \(\sigma_F = \sigma_\uparrow + \sigma_\downarrow\), and \(\sigma_\uparrow, \sigma_\downarrow\) are the bulk conductivities of the ferromagnet. We note that in the semi-infinite ferromagnet \(\Delta \zeta_F(x) = C \exp(x/L_F)\), where \(L_F\) is the ferromagnet spin diffusion length. This implies that \(\Delta \zeta_F(0) = \Delta \zeta_F(0)/L_F\). Also \(\Delta \zeta_F(0) = -\tanh(w/L_N)\Delta \zeta_N(0)/L_N\). Substituting these formulas in Eqs (3) and (4), eliminating \(\Delta \zeta_F(0)\), and using Eq. (1) we finally obtain:
\[ \gamma(0) = \frac{\gamma_c r_c + \gamma_F r_F}{r_F + r_c + r_N / \tanh(w/L_N)}, \]
and
\[ \Delta \zeta_N(0) = \frac{2j[\gamma_c r_c + \gamma_F r_F] r_N}{(r_c + r_F) \tanh(w/L_N) + r_N}. \]

Here we introduced the resistances \(r_F = L_F/\sigma_F/(4\sigma_\uparrow\sigma_\downarrow)\), and \(r_N = L_N/\sigma_N\). Using Eqs. (2) and (6) we can calculate the spin polarization at the N/S interface, \(P = P(w) \propto \Delta \zeta_N(w)\), which yields:
\[ P(w) = \frac{P_0}{\kappa \sinh(w/L_N) + \cosh(w/L_N)}, \]

where \(\kappa = (r_c + r_F)/r_N\) and \(P_0 \propto \gamma_c(r_c/r_N) + \gamma_F(r_F/r_N)\), is the limiting value of the spin polarization at small \(w\). The results of our fitting procedure are shown in Fig 3 with \(L_N \approx 285\) nm and \(\kappa \approx 3.5\).

The qualitative dependence of \(P/P_0\) for three different values of \(\kappa\) is shown in Fig. 4. As can be seen from the plot, the thickness dependence of \(P\) is much sharper than the simple exponential dependence, \(P \propto \exp(-w/L_N)\), which is often used to fit the spin diffusion data. Indeed, at small \(w\), \(P \approx 1 - \kappa w/L_N\) rather than \(1 - w/L_N\). It means that for \(\kappa > 1\) the spin polarization in Eq. (7) decays faster than the simple exponent. Thus, if we attempted to fit our data with a simple exponential dependence we would obtain \(L_{\text{eff}} \approx L_N/\kappa\). In our case, this is about three times smaller than the actual value. The best fit with the simple exponential dependence gives \(L_{\text{eff}} \approx 130\) nm (see Fig. 3). In addition, a naive interpretation would give different values of the apparent spin diffusion length for different ferromagnetic spin injectors and F/N interfaces of different quality, which is obviously a non-physical result.

Eq. (7) is valid at low temperatures when Andreev reflection dominates the transport across the interface. At higher temperatures we have to take into account the thermally activated tunneling of quasiparticles, which
leads to a non-zero spin current at the N/S interface. Following Takahashi et al. [23] let us introduce the (spin-independent in our case) tunnel conductance for the N/S interface \( \Sigma_{NS} = \Sigma_N \chi(T) \) where \( \Sigma_N \) is the tunnel conductance between the two normal metals (i.e. above the superconductivity threshold \( T_c \)) and \( \chi(T) \) is the so-called Yosida function [23] describing increase of the tunneling conductance as the temperature rises from 0 to \( T_c \).

\[
\chi(T) = 2 \int_{\Delta}^{\infty} \frac{E_k}{\sqrt{E_k^2 - \Delta^2}} \left( - \frac{\partial f_0}{\partial E_k} \right) dE_k, \quad (8)
\]

where \( f_0(E_k) \) is the Fermi distribution function and \( E_k = \sqrt{\xi_k^2 + \Delta^2} \) is the quasi-particle energy with \( \xi_k \) being a one-electron energy relative to the chemical potential of the superconductor.

In the absence of the spin-flip transition at the N/S interface and in S-region the boundary condition \( \gamma(w) = 0 \) has to be replaced with [22, 23]

\[
2 j \gamma(w) = -\Sigma_N \chi(T) \Delta \zeta(w) \quad (9)
\]

Using the boundary condition [3] we can repeat the above calculations and obtain:

\[
P(w, T) = \frac{P_0 [1 + \kappa \mu \chi(T)]^{-1}}{g(T) \sinh(w/L_N) + \cosh(w/L_N)}, \quad (10)
\]

where \( \mu = r_N \Sigma_N \) and

\[
g(T) = \frac{\kappa + \mu \chi(T)}{1 + \kappa \mu \chi(T)} \quad (11)
\]

Since \( \chi(T) \) strongly depends on the temperature both the maximum value and the shape of \( P(w) \) strongly depend on the temperature. A typical temperature dependence of the spin polarization described by Eq. (10) is shown in Fig. 5. If, as previously, we attempt to interpret Eq. (10) using a simple exponential dependence \( P \propto \exp(-w/L_{eff}(T)) \) we will get a spurious temperature dependence of the apparent spin-diffusion length \( L_{eff}(T) \) (see Fig. 5), as was inferred by Geresdi et al. [24], demonstrating that neglecting the backflow effect could lead to erroneous results.

\[
\begin{align*}
\text{FIG. 4: Normalized spin polarization } P/P_0 \text{ for different values of } \kappa. \\
\text{FIG. 5: Spin polarization } P(w, T)/P_0 \text{ (Eq. (10)) for } \kappa = 3.5 \text{ and } \mu = 3.
\end{align*}
\]

We use several approximations in our description of the experimental geometry, such as adopting a one dimensional model for what is a 3D problem and using boundary conditions at the N/S interface that assume only Andreev reflection below the gap, hence neglecting processes above the gap. While these approximations may introduce some systematic errors, they are unlikely to significantly affect the rate of spin polarization decay, which determines the values of spin diffusion length. We also note that within the same approximations, it is possible to obtain a complete set of data needed for the determination of spin diffusion length from a single sample by sequentially positioning the tip for PCAR measurements along the side of the normal electrode, as shown.
In summary, the backflow effect on spin diffusion and spin accumulation is formulated as a consequence of preferential majority scattering near normal metal - superconducting interface. It is found that spin current probed by Andreev Reflection measurements gradually decays, as we increase the thickness of the normal layer, revealing the scale of spin diffusion in the normal metal. The measured spin diffusion length in gold of approximately 285 nm, more than two time larger than that one would have obtained using a simple exponential fit. While our experimental results are described specifically for a normal metal - superconducting interface, we emphasize the role of boundary conditions, noting that qualitatively similar effects would take place for normal metal - ferromagnetic interface as well, and thus are relevant for other spin diffusion length measurement techniques.

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