Material-specific spin filtering in ferromagnet/superconductor ballistic nanojunctions

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We study spin-dependent electronic transport across ferromagnet/superconductor ballistic junctions modeled using tight-binding Hamiltonians with s, p and d orbitals and material-specific parameters. We show that by accurately modeling the band structure of the bulk materials, one can reproduce the measured differential conductance of Cu/Pb nanocontacts [1,2]. In contrast the differential conductance of Co/Pb contacts can only be reproduced if an enhanced magnetic moment is present at the interface.

During the last few years numerous experimental studies of electronic transport properties of nanostructures containing both ferromagnets (F) and superconductors (S) have been reported [1–11]. Such structures exhibit novel features, not present in normal-metal/superconductor (N/S) junctions, due to the suppression of electron-hole correlations in a ferromagnet when a large exchange field is present. Spin-dependent transport in structures containing magnetic materials is also underpinning technological advances in spintronics, where magnetic materials are used as spin-filters. A key parameter is the degree of polarization \( P \) of the current in a ferromagnet, which is currently the subject of an intense debate (see, for example [12–15]). In the two spin fluid approach \( P \) is defined as:

\[
P = \frac{I^\uparrow - I^\downarrow}{I^\uparrow + I^\downarrow},
\]

where \( I^\uparrow (I^\downarrow) \) is the current carried by spin-up (spin-down) electrons. Unfortunately \( I^\uparrow \) and \( I^\downarrow \) cannot be measured separately in an isolated ferromagnet and therefore \( P \) cannot be determined directly. As Tedrow and Meservey showed in references [16–19], \( P \) can be estimated by attaching F to a superconductor through a tunnel junction and taking advantage of the superconducting gap in the density of states (DOS) of the superconductor. This method, however, has a limitation, namely that the insulating layer has to be uniform, which is a difficult situation to reach for many ferromagnetic materials. In particular atomic size pin-holes can short-circuit most of the tunneling current [20], giving rise to spurious \( I-V \) tunneling curves.

To overcome this problem, an alternative method has been proposed [21] which exploits the suppression of Andreev reflection at F/S ballistic junctions. In this Letter we argue that an understanding of spin-polarized transport in such hybrid nanostructures requires an understanding of surface scattering which goes beyond the heuristic analysis of references [1–3]. We present detailed calculations of the conductance of Co/Pb and Cu/Pb ballistic interfaces, which show that although the experiments of references [1,2] tell us little about bulk magnetization, they do provide the first evidence of enhanced surface magnetization at the Co/Pb interface.

Providing that S is much longer than the superconducting coherence length, the sub-gap conductance of a F/S junction is solely determined by Andreev reflection at the interface. The idea used in references [1–3] to estimate the polarization is based on the fact that, in the absence of spin-flip processes, as \( P \) is increased, Andreev reflection is suppressed in favor of normal reflection. In the present calculation, F/S and N/S junctions are described using a tight-binding Hamiltonian on a \( fcc \) lattice with hopping to first nearest neighbors. In order to accurately reproduce the band structure of real materials, we take into account 9 orbitals per site (s, p and d) and calculate the tight-binding parameters by fitting the band structure obtained from density functional calculations [22]. The fit is made using OXON [22], a tight-binding code which minimizes the deviation between the LDA results and dispersion curves obtained from the tight-binding parameterization. As reference points in

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the band structure, we take the eigenvalues at four high symmetry points in the \textit{fcc} Brillouin zone (namely \Gamma, L, X and W) of each band. Moreover we further checked the symmetry of the resulting tight-binding bands along several directions in the Brillouin zone. It is worth noting that, in order to get a good fit for both majority and minority electrons of Co, the band structures of the different spin species are fitted separately as if they were different materials.

The junction is modeled by coupling a ferromagnetic semi-infinite lead on the left-hand-side to a superconducting semi-infinite lead on the right-hand-side, using an interface Hamiltonian $H_{\text{int}}$. The hopping matrix elements are chosen to be the mean square of the bulk elements, with a sign equal to that of the largest of the two bulk parameters.\(^1\) The conductance of the junction is evaluated within the scattering approach outlined in \cite{23} where transport amplitudes are calculated using a recursive Green’s function technique.

Since we consider only clean interfaces the system is translationally invariant in the directions parallel to the interface, so that the total scattering coefficients are given by the sum over all possible Bloch wave-vectors in the 2-dimensional Brillouin zone. In our calculations we take the interface perpendicular to the (110) direction and sum over 900 Bloch wave-vectors, which corresponds to a junction diameter of the order of the experimental one (junction area $\sim 10 \text{ nm}^2$). In order to make a comparison with the experimental data \cite{12}, we define the following dimensionless quantity:

$$g(V) = \frac{G_S(V) - G_N(V)}{G_N(0)}, \quad (2)$$

where $G_S(V)$ ($G_N(V)$) is the differential conductance at voltage $V$, when the S-lead is in the superconducting (normal) state.

In Fig. 1 the computed $g(V)$ curves are plotted for Cu/Pb and Co/Pb junctions at $T=4.2 \text{ K}$ using the superconducting gap for bulk Pb ($\Delta = 1.26 \text{ meV}$). Fig. 4 shows that the measured $g(V)$ curve of references \cite{12,13} is well reproduced for Cu/Pb, but in the absence of surface magnetism, the Co/Pb result disagrees with experiment. At present there exist no ab-initio calculations of the Co/Pb interface, mainly due to difficulties associated with modeling heavy elements such as Pb. Furthermore little is known experimentally about surface magnetism at this important interface. Nevertheless it is known that related interfaces can yield surprises. For example in recent experiments involving F/S multilayers and F/S trilayers both the presence and absence of magnetically dead monolayers at the surface, when S is in the normal state, have been reported depending on the material and geometry (see for example \cite{24,25}). In addition, an enhanced magnetic moment has been found in ferromagnetic clusters, isolated or deposited onto a film. For example in experiments on Co, Ni and Fe clusters \cite{27–29}, the magnetic moment was found to increase up to 36% higher than the bulk value. LMTO calculations of Co islands grown on Cu films \cite{30} also show an increase of about 40% in the local spin polarization. This enhancement of the magnetic moment of a transition metal at an interface is mainly due to the suppression of the quenching of the orbital component of the magnetization \cite{31}. In bulk magnetic transition metals the orbital component of the magnetization is strongly suppressed by the cubic crystal field. In contrast at an interface the crystal symmetry is broken and the quenching is only partial. This leads also to an enhancement of the spin component which is strongly spin-orbit coupled to the orbital one. Finally, for F/S interfaces a decrease of about 10% in the average magnetic moment in Fe has been reported in Fe/Nb bilayers while cooling the sample through the superconducting critical temperature \cite{32}. This evidence has been explained \cite{33} by the presence of a cryptoferromagnetic state within islands of reduced exchange field in the Fe layer. The phenomenon of cryptoferromagnetism \cite{24} consists of the formation of a small-scale domain structure within a ferromagnet in the vicinity of a F/S interface. In general, however, the cryptoferromagnetic state in both samples is possible only in the case of weak ferromagnets, such as Gd \cite{33}. No such behavior has yet been predicted or observed for Co.

\(^1\)It should be noted that other choices have been considered in the literature, including the geometric mean using the above sign rule and the geometric mean with a sign equal to that of the product of the bulk parameters. We have repeated the calculations of this Letter for both of these choices and find that neither is capable of reproducing the experimental results for both Co/Pb and Cu/Pb.
In the case of references [1,2], an altered surface magnetization in Co could be produced by the peculiar geometry of the sample: the portion of Co in contact with Pb has an approximate area of $(10 \, \text{nm})^2$, in which one domain can fit. This suggests that the exchange field at the interface can be larger than the exchange field in the bulk. In what follows, we show that an increased exchange field at the interface does indeed yield good agreement with the experimental $g(V)$ curve. Fig. 2 shows the calculated zero bias, zero temperature conductances $G_S$ and $G_N$, for the Co/Pb junction in the presence of a single surface monolayer of Co with arbitrary exchange field $h$ entering all the orbitals. This shows that there exists a range of values of the surface magnetization $h$ for which $G_S > G_N$ (in agreement with experiment), with the largest $g(0)$ found for $h = 1.84 \, \text{eV}$. We also considered the possibility of a tilted magnetization in the inserted monolayer with respect to the bulk magnetization, but as a general feature, we find that for small angles $\phi$, $G_S$ does not vary much and thereafter it decreases (see Fig. 3). In Fig. 4 the $g(V)$ curve for Co/Pb is shown at 4.2 K for $h = 1.6 \, \text{eV}$, parallel to the bulk magnetization. This closely matches the experimental plot in reference [2] and demonstrates that the experimental results for $g(V)$ of both Cu/Pb and Co/Pb junctions can be reproduced, provided one accounts for additional surface magnetism in the Co.
FIG. 4. $g(V)$ curve for Co/Pb obtained from the spd-model when the exchange field in the Co monolayer at the interface is $h = 1.6$ eV.

Finally we conclude by discussing the difference between the polarization $P$ of a bulk ferromagnet and the polarization $P$ of a F/N interface made from the same ferromagnet. Since the dependence of the transmission coefficients on the energy is small (around 1%) in the range we are considering here, we focus on the zero bias, zero temperature limit. For a F/N interface the definition of $P$ given by the equation (1) becomes

$$
P = \frac{G^\uparrow - G^\downarrow}{G^\uparrow + G^\downarrow},$$

where $G^\uparrow (G^\downarrow)$ is the conductance for majority (minority) electrons in units of $\frac{e^2}{h}$ normalized to the corresponding number of open channels. From the results obtained using the spd-model we find that while the polarization of bulk Co is negative ($P_{\text{Co}} = -0.400$), the polarization of a Co/Pb(N) junction is positive (for instance, $P_{\text{Co/Pb}} = +0.400$ when $h$ at the interface equals the bulk value and $P_{\text{Co/Pb}} = +0.275$ when at the interface $h = 1.6$ eV). This striking difference occurs, because in the former case $P$ is determined solely by the DOS, with the minority electrons possessing a larger DOS (mainly d-like) than the majority electrons DOS (s,p,-d-like) [23]. In contrast for a Co/Pb(N) junction, $P$ is also determined by the mismatch between the band structures of the two materials. In this case, despite their large DOS, minority electrons of Co are more strongly scattered at the interface with Pb (whose DOS is mainly s- and p-like) than Co majority electrons. This makes clear that in general the polarization of a F/N junction also depends strongly on the band structure of the non-magnetic material. As a further example, we have also computed transport properties across an Ir-Co interface. Assuming bulk magnetization at the interface we obtain $P_{\text{Co/Ir}} = -0.010$, which has the opposite sign with respect to Co/Pb. This arises since the DOS at the Fermi energy of Ir is mainly d-like and the mismatch of the band structures with Co is larger for majority than for minority electrons.

In conclusion we have shown that a detailed description of the band structures of the individual materials and of the interface is needed to accurately describe the I-V curves of S/F ballistic junctions. In particular we have demonstrated that band structure mismatch of the two materials can give rise to a polarization of the whole junction which is completely different from the bulk polarization of the ferromagnetic. This casts some doubt on the reliability of simple models based solely on surface scattering to describe such junctions. Finally we found that for Co/Pb junctions the experimental I-V curves are well reproduced if an enhancement of the magnetization of Co at the interface is assumed. This is consistent with the reduction of the quenching of the orbital component of the magnetic moment of a ferromagnetic transition metal at an interface, as reported recently in literature.

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