Point-Contact Andreev Reflection in Ferromagnet/Superconductor Ballistic Nanojunctions

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A critical analysis of point-contact Andreev reflection (PCAR) in ferromagnet/superconductor ballistic junctions is presented, based on tight-binding Hamiltonians with s, p and d orbitals and material-specific parameters. By accurately modeling the band structure of the bulk materials, we show that to reproduce the measured differential conductance of Cu/Pb and Co/Pb nanocontacts one needs a detailed understanding of interface parameters which goes beyond simple averages of bulk matrix elements. As an example of how such a modification of surface parameters can affect spin-polarized transport, we explore the effect of an enhancement of the surface magnetization and show that this can significantly improve agreement with experiment. We also demonstrate that in general the spin polarization $P$ of the junction may be different from the bulk polarization $P$ of the magnetic metal used. For bulk Co we find $P = -0.4$, whereas for the Co/Pb interface $P = +0.4$ and for the Co/Ir interface $P = -0.01$.

**Keywords:** Nanojunctions, Ferromagnets, Superconductors, Andreev Reflection.

In the last decade experimental studies of electronic transport properties of nanostructures containing both ferromagnets ($F$) and superconductors ($S$) have received increasing attention. 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 defined as:

$$P = \frac{I^+ - I^-}{I^+ + I^-},$$  \hspace{1cm} (1)

where $I^+$ and $I^-$ cannot be measured separately in an isolated ferromagnet and therefore $P$ cannot be determined directly. As Tedrow and Meservey showed, $P$ 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, giving rise to spurious $I$-$V$ tunneling curves.

An alternative method based on point-contact Andreev reflection (PCAR) has been proposed which exploits the suppression of Andreev reflection at $F/S$ ballistic junctions. This method has high energy resolution, avoids the need of applied magnetic fields and has no restrictions on the sample geometry. During the last six years PCAR has been extensively used in experiments for determining the polarization of hard ferromagnets (Co, Ni, Fe), permalloy ($\text{Ni}_x\text{Fe}_{1-x}$), and conducting ferromagnetic oxides ($\text{SrRuO}_3$, $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$). However the deduction of $P$ from a PCAR measurement presents difficulties due to the various fitting parameters entering the models used for analysing the data, and the problem of determining the transport regime (i.e., ballistic, diffusive...
or intermediate). These issues are currently the subject of an intense debate.17, 20, 23–27

In this paper we present a critical analysis of PCAR by introducing a tight-binding description which correctly describes the band structure of the bulk materials. Our model goes far beyond the heuristic analysis.1, 2, 16 We present detailed calculations of the conductance of Co/Pb and Cu/Pb ballistic interfaces, which show that the polarization of the current measured in the experiments1, 2 tells us little about bulk magnetization, but may provide evidence of large spin-selective interfacial scattering.

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 used1, 2, 16 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.25 The fit is made using OXON,29 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 the band structure, we take the eigenvalues at four high symmetry points in the fcc Brillouin zone (namely Γ, 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, using the fitting procedure published.30 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. Superconductivity in Pb is modeled by introducing an orbital-independent superconducting order parameter Δ which couples particle and hole degrees of freedom on each site (see Ref. [31]). Note that the exchange coupling in magnetic transition metals is typically three order of magnitudes larger than the superconducting gap in Pb. For this reason we do not expect the spin-polarization of the junction to change with the onset of the superconductivity, and therefore the same tight-binding parameters can be used in the normal and superconducting case. In view of the large electron density at the Fermi energy in metals and the resultant screening of the Coulombic potential, we do not expect significant band-bending at the interface and therefore we simply align the Fermi energies of the two bulk 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. The conductance of the junction is evaluated within the scattering approach outlined15, 35 where transport amplitudes are calculated using a Green’s function technique.

We assume clean interfaces so that the system is translationally invariant in the directions parallel to the interface, and 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 sum over 900 Bloch wave-vectors, which corresponds to a junction diameter of the order of the experimental one (junction area ~10 nm$^2$).

For definiteness we take the direction of growth to be the (110), i.e., the interface is oriented perpendicular to the (110) direction. A different choice of crystal direction, however, is not expected to yield an appreciable difference in the results. The transport in bulk Cu (or Pb or Co) is very weakly dependent on the crystalline orientation and the only dependence should arise at the interface with a different material. However we are using a rigid tight-binding model, where charge rearrangement is not considered. This means that within our model differently oriented interfaces between uneven metals should not differ appreciably. In order to make a comparison with the experimental data,1, 2 we define the following dimensionless quantity:

$$g(V) = \frac{G_\text{S}(V) - G_\text{N}(V)}{G_\text{N}(0)},$$

where $G_\text{S}(V)$ ($G_\text{N}(V)$) is the differential conductance at voltage $V$,2 when the $S$-lead is in the superconducting (normal) state.

The assumption of clean interfaces is suggested by the experiments,1, 2 because of the reported small sample-to-sample variations of $g(V)$. Therefore in what follows, the surface hopping parameters at the interface are chosen to be an average of the corresponding bulk parameters.

In Figure 1 the computed $g(V)$ curves are plotted for Cu/Pb and Co/Pb junctions at $T = 4.2$ K using the superconducting gap for bulk Pb ($\Delta = 1.26$ meV). In the bias voltage range [0, 2] mV, Figure 1 shows that while the measured $g(V)$ curve1, 2 is quite well reproduced for Cu/Pb (note that we have no free adjustable parameters), the Co/Pb result disagrees with experiment. As noted, the measured $g(V)$ for large biases takes finite values, deviating from the expected behaviour.1 This deviation is most visible for Cu/Pb junctions and it is attributed either to strong coupling effects in Pb or to the partial gap suppression due to the high current density achieved in the experiment. In this paper we do not address this issue and we concentrate on the range of bias voltages [0, 2] mV, where our approach is appropriate.

At present there is no ab-initio systematic study of the Co/Pb interface, hence it is difficult to provide an accurate.

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description at the atomic scale. In view of this uncertainty we have considered alternative choices of surface parameters, 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 paper for both of these choices and find that neither is capable of reproducing the experimental results for both Co/Pb and Cu/Pb. In studies of magnetic activity of monolayer grown on substrates, discrepancies between theoretical models and experiments have been found and ascribed to the poor quality of the deposited film (see for example in Refs. [37–39]). However, the absence of sample-to-sample fluctuations suggests that disorder at the interface cannot explain the disagreement. Furthermore, recent density functional calculations for the Co/Pb interface, show very similar \( I-V \) curves to those presented here. In that case chemical disorder at the interface is introduced by the peculiar geometry of the deposited film. As noted, the measured \( g(V) \) for large biases takes finite values, deviating from the expected behavior. This deviation is most visible for Cu/Pb junctions and it is attributed to strong coupling effects in Pb or to the partial gap suppression due to the high current density achieved in the experiment.

Fig. 1. Comparison between the \( g(V) \) curves obtained using the spd-model and the experimental data. The dashed line is the theoretical curve relative to the Co/Pb junction, while the squares represent the experimental data. As noted, the measured \( g(V) \) for large biases takes finite values, deviating from the expected behavior. This deviation is most visible for Cu/Pb junctions and it is attributed to strong coupling effects in Pb or to the partial gap suppression due to the high current density achieved in the experiment.

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. 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. This evidence has been explained by the presence of a cryptoferromagnetic state within islands of reduced exchange field in the Fe layer. The phenomenon of cryptoferromagnetism 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. No such behavior has yet been predicted or observed for Co.

In the case of 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 (i.e., in the last monolayer of Co in contact with Pb) does indeed yield good agreement with the experimental $g(V)$ curve. Figure 2 shows the calculated zero bias, zero temperature conductances $G_x$ and $G_S$, for the Co/Pb junction when a single monolayer of Co, with arbitrary exchange field $h$ entering all the orbitals, is introduced at the interface between Co and Pb leads. This shows that there exists a range of values of the surface magnetization $h$ for which $G_x > G_N$ (in agreement with experiment), with the largest $g(0)$ found for $h = 1.84$ 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 $\phi$, $G_x$ does not vary much and thereafter it decreases (see Fig. 3). This is an expected result: at $\phi = \pi$, in fact, $G_x$ has to be smaller than the conductance at $\phi = 0$, because the former case corresponds, essentially, to the introduction of an additional “interface.” In addition we find that for all values of $\phi$, $G_x$ decreases more rapidly than $G_N$ with increasing $\phi$. Again this is expected, since the Andreev reflection coefficient decreases as roughly the square of the transmission coefficient, whereas $G_N$ decreases only linearly. However, it is worth noting that for all values of $h$, $G_x$ is not a smooth function of $\phi$ due to complicated interference effects (remember that we are dealing with 18 orbitals per site per spin) induced by the presence of this “interface.” For $h = 1.6$ eV, for example, such interference effects produce a pronounced resonance determined by the details of the Hamiltonian. Of course, marked resonances are also present for other intermediate values of $h$. In Figure 4 the $g(V)$ curve for Co/Pb is shown at 4.2 K for $h = 1.6$ eV, parallel to the bulk magnetization. This quite closely matches the experimental plot and shows 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.

Whether or not an enhanced magnetization at the surface actually occurs, the above analysis demonstrates that magnetic structure of the interface is a crucial factor in explaining experimental results and an altered interface magnetization can improve the agreement between theory and experiment.

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 Eq. (1) becomes

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

(3)

where $G^+(G^-)$ is the conductance for majority (minority) electrons in units of $e^2/h$. 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). 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 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 stated the 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. As expected, we have demonstrated with actual examples (Co/Pb and Co/Ir junctions) 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 ferromagnet. This casts some doubt on the reliability of simple models based solely on surface scattering to describe such junctions. In addition we found that for Co/Pb junctions the experimental $I$-$V$ curves can be reproduced quite well if a large enhancement of the magnetization of Co at the interface is assumed.

Our calculations show that the polarization of the current is extremely sensitive to surface parameters. Ab-initio calculations would, in principle, provide a definitive answer to the problem of determining spin-dependent transport properties in Co/Pb junctions. In practice, however, this would require the exact knowledge of the geometry of the interface, which is not currently available. In this paper we have adopted a more viable semi-empirical approach, whereby the bulk band structures are accurately described and the interface parameters are given by averages of the corresponding bulk parameters. We believe that a first principle model of the interface would be of great interest for the future and that the present calculations can be an incentive for further investigation in this direction.

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References

29. OXON (the Oxford O(N) tight binding code) was developed at the Materials Modelling Laboratory of the Department of Materials at the University of Oxford.
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