Origin of spin-polarization decay in point-contact Andreev reflection

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We have performed point contact Andreev reflection measurements with a view to study the correlation between measured spin polarization and the interface scattering parameter Z extracted from fits of the modified Blonder-Tinkham-Klapwijk model to the conductance-voltage curves of superconductor/ferromagnet point contacts. A simple model describing spin-flip scattering in the interface region identifies Z^2 as the effective scattering parameter and predicts that the spin polarization decays exponentially with Z^2, in agreement with experimental data.

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A spin-polarized current is the essential ingredient for transport phenomena currently exploited in, for example, magnetic read head and nonvolatile memory devices.[1] It originates from an imbalance in the spin-up and spin-down electron contributions to the current in a ferromagnet. This spin polarization, not to be confused with the magnetization of the ferromagnet concerns the conduction electrons at the Fermi level.

Measurement techniques which directly study spin polarization contribute to the understanding of spin-polarized transport phenomena as well as the development and improvement of spin-electronic devices. A well-known technique is spin-polarized tunneling (SPT),[2] pioneered by Meservey and Tedrow in the 1970's, which measures the spin polarization of the tunneling electrons in a superconductor/insulator/normal metal tunnel junction. During the last five years, point contact Andreev reflection (PCAR) has emerged as a new approach for direct measurement of spin polarization.[3–8] This technique involves transport through a superconductor/normal metal (S/N) point contact in which the superconductor serves as the probe for the spin polarization. The Andreev reflection process[9] allows electrons to cross the S/N interface at energies in the superconducting band gap. Basically, in this process two electrons from the N electrode with opposite spin enter the S electrode by forming a Cooper pair. The Andreev reflection process cannot occur when N is fully spin polarized, since no Cooper pairs can be formed, and accordingly the degree of spin polarization influences the conductance of the contact. Based on the character of the electrons participating in the transport processes, the spin polarization probed with PCAR is predicted to be different from the tunneling spin polarization probed with SPT.[10] In this respect, PCAR can be regarded as a method complementary to SPT.

The transport in S/N contacts, in which N is a nonmagnetic metal, was originally described by the Blonder-Tinkham-Klapwijk (BTK) model.[11] Interface transparency is accounted for by a planar δ-potential with dimensionless strength Z. The BTK model has been modified[4,6] to describe S/N contacts in which N is a magnetic metal and has been used to extract the spin polarization P by fitting the model to conductance-voltage (dI/dV-V) curves. It has been observed that P is systematically suppressed with decreasing interface transparency,[6,8] i.e., increasing Z, and empirically described with a polynomial which is second order in Z. Although the extrapolation of P towards small Z has been used to estimate the intrinsic spin polarization of magnetic materials, the physical mechanism behind this spin polarization decay has not been addressed. In this paper, we present PCAR measurements on Co and Fe for a wider range of interface transparencies than has been previously reported, from which a universal exponential decay of P as a function of Z^2 can be deduced. A simple model, incorporating spin-flip scattering in the interface region of the contact, identifies Z^2 as an effective scattering parameter and explains the decay of P. We further substantiate the model by presenting PCAR measurements on the magnetic rare-earth metal Gd and by considering existing data on half-metallic CrO_2.

Our contacts are obtained by pressing a superconducting tip onto a sample by means of a mechanically driven mechanism. This experimental approach is similar to what is used in earlier work.[4,6–8] Conductance (dI/dV) is measured with a standard lock-in technique at liquid helium temperatures. Both Nb and Pb are used as superconducting tips. The samples are either UHV sputter-deposited films of 50 nm thickness or bulk samples with a mechanically polished surface. Tip and sample are brought into physical contact while immersed in liquid helium. In general, changes in contact resistance and Z are obtained by applying a short voltage pulse or due to mechanical drift over a time scale long compared to the measurement time.

Before we address the P-Z correlation observed for magnetic metals, we show measurements performed on nonmagnetic metals to demonstrate the accuracy of the modified BTK model and the reliability of our contacts. Figure 1 shows representative examples of a Nb/Cu and Pb/Al contact measured at 4.2 K. The conductance is normalized by its value at 7 mV and has conductance maxima marking the edge of the superconducting band gap. Generally, the measured superconducting band gap is close to the bulk value, and, most importantly, the P obtained from a least squares fit of the modified BTK model (solid lines) is zero. We emphasize that a best fit is obtained with a fitted temperature T_f systematically about 1 K higher than the experimental temperature. This additional broadening cannot be caused by Joule heating, since T_f is approximately constant in a range

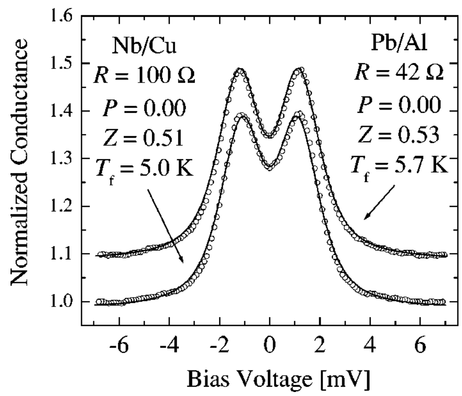

FIG. 1. Test measurements at 4.2 K on the nonmagnetic elements Cu and Al. The solid lines are fits of the modified BTK model. The curves have a vertical offset for clarity.

in contact resistance from Ω's to kΩ's. The extra broadening might be caused by the proximity effect,[12] not accounted for by the BTK model. This would be consistent with calculations[13] showing increased broadening of the conductance curve when the proximity effect is included.

For the ferromagnetic transition metals, Strijkers *et al.*[6] reported a systematic suppression of P with Z for $Z<0.4$. The measured polarization was plotted as a function of Z and an empirical second order polynomial was used to fit the results. We have measured the spin polarization of the ferromagnetic metals Co and Fe for Z values up to 0.9. Two representative measurements of a Pb/Co contact are shown in Fig. 2. The P extracted from the fit (solid line) is larger for the contact with the lower Z value. We observed no correlation between P and contact resistance, implying that the Z value alone is decisive in determining the magnitude of P. The inset of Fig. 3 shows P as a function of Z obtained for Co. This data essentially reproduces the results of Strijkers *et al.* However, a second order polynomial in Z (solid line) does not give a convincing fit for $Z>0.4$. For purposes which become clear below, we present in Fig. 3 our Co and Fe data as a function of Z^2 rather than Z, revealing an exponential-like decay in P. Apart from the considerable

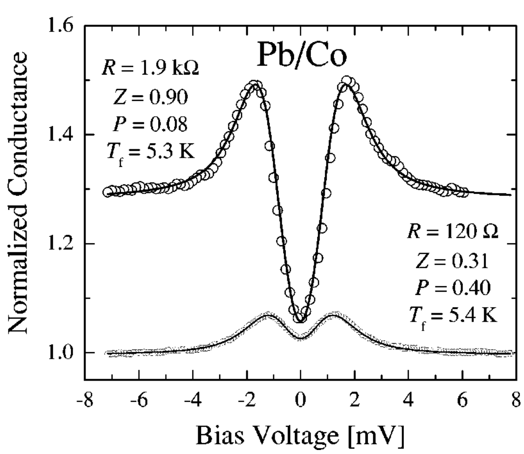

FIG. 2. Two Pb/Co contacts with different Z-values measured at 4.2 K with BTK fits (solid lines). The curves have a vertical offset for clarity.

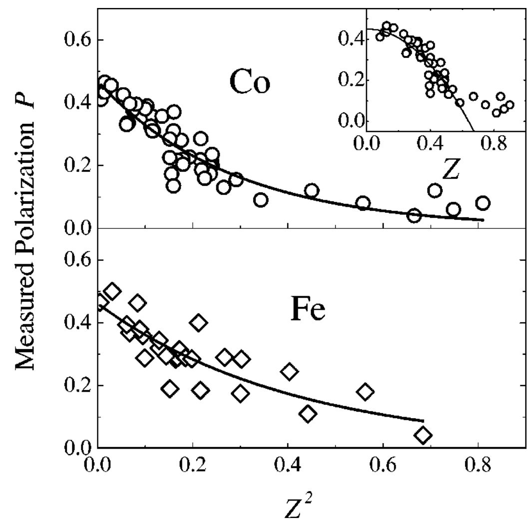

FIG. 3. P plotted as a function of Z^2 for Co and Fe. The solid lines are fits to an exponential decay. The inset shows P as a function of Z for Co together with a polynomial fit of second order.

spread in the data, which is comparable to what is reported by Strijkers *et al.*, the exponential correlation is reproduced using multiple tip and sample combinations. We do not resolve a dependence on the choice of Pb or Nb as superconductor or the choice of a 50 nm thin film vs a bulk sample.

The difficulty in understanding the correlation between measured polarization and Z^2 lies in the fact that Z is not a direct physical quantity, but rather, it is the strength of a δ potential which in reality does not exist. In order to understand the experimental results, Z^2 must be identified with a physical mechanism responsible for elastic electron scattering. Three potential contributions can be distinguished: scattering due to the mismatch of the electronic band structures at the interface, specular reflection at a tunnel barrier, and scattering at impurities and lattice defects. The first contribution to Z^2 is intrinsic since it is present even for a perfectly clean contact without defects. Based on a free-electron approximation,[14] this contribution has an order of magnitude of 10^{-2}. An *ab initio* calculation,[15] fully accounting for the transition metal band structure, results in an intrinsic mismatch contribution of roughly 10^{-1}. These numbers are small compared to the experimental Z^2 values, suggesting that the mismatch plays a relatively unimportant role in our contacts. A tunnel barrier resulting from a single monolayer of typical insulating oxides, would be equivalent to a δ-potential barrier with $Z^2>10$, which is much larger than the experimental values. Therefore, we consider the presence of a tunnel barrier at the interface rather unlikely, and we assume that mostly impurity scattering and scattering at lattice defects contribute to Z^2.

By modeling this type of interface scattering with the use of a δ potential, as is done in the BTK model, one is assuming a picture in which an incoming electron is reflected or transmitted as a result of at most a single scattering event [Fig. 4(a)]. In practice, however, an electron may be transmitted or reflected as a result of multiple scattering events in forward and backward direction in an extended scattering region [Fig. 4(b)]. Accordingly, the Z^2 value obtained from

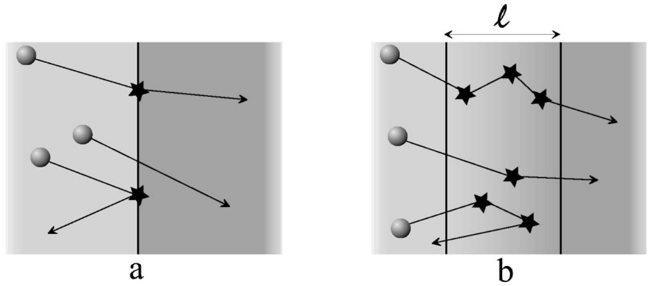

FIG. 4. Scattering at a specular interface modeled by a planar δ potential (a) and scattering in an extended interface region of width l (b).

fitting the BTK model to our contacts can be considered as an ''effective'' scattering parameter which measures multiple scattering events. This can be modeled simply by calculating the transmission T of the extended scattering region and comparing the result with the transmission of the δ potential. The problem of calculating the transmission in the extended scattering region is related to a random walk problem. If l is the width of the scattering region, λ the electron mean free path, and ψ a scattering anisotropy defined by the ratio between the probability for forward and backward scattering, the solution is

$$T=\frac{1}{1+\frac{1}{1+\psi}\frac{l}{\lambda}}. \tag{1}$$

This expression has the same form as the transmission for the δ potential,[11] which reads

$$T=\frac{1}{1+Z^2}. \tag{2}$$

Comparison of the two expressions for T suggests that

$$Z^2=\frac{1}{1+\psi}\ \frac{l}{\lambda}. \tag{3}$$

The quantity Z^2 scales with l/λ, which is a measure for the average number of scattering events of a transmitted electron. If the electrons scatter mostly in forward direction, ψ is large and Z^2 is small. In the fully backward scattering limit, ψ is zero and $Z^2=1l/\lambda$.

In addition to a quantitative expression for Z^2, the model provides a relation between the spin polarization of transmitted electrons and Z^2. To this end, we assume that the scattering region is nonmagnetic and that for each scattering event there is a spin-flip probability α. The transmitted polarization can be calculated analytically using statistical analysis similar to that used in obtaining Eq. (1). If P_0 is the spin polarization of the incoming electrons before the scattering region, the exact result is

$$P=P_0\frac{(1+Z^2)\,\eta}{(1+2\alpha\psi)\sinh(\eta Z^2)+\eta\cosh(\eta Z^2)}, \tag{4}$$

TABLE I. Fit results (see text for details).

Material	P_0	$\alpha\psi$
Co	0.47 ± 0.02	1.8 ± 0.1
Fe	0.46 ± 0.03	1.2 ± 0.2
Gd	0.45 ± 0.04	2.1 ± 0.3
CrO_2 (Ref. 8)	0.96 ± 0.02	0.16 ± 0.03

where $\eta^2=4\alpha(1+\psi)+4\alpha^2(\psi^2-1)$. For large ψ, i.e., dominant forward scattering, the dependence on Z^2 in the above expression reduces essentially to an exponential decay

$$P\approx P_0\exp(-2\alpha\psi Z^2). \tag{5}$$

This result is similar to what is observed in the experiment. The solid lines in Fig. 3 are fits of Eq. (5) and the fitting parameters P_0 and $\alpha\psi$ are listed in Table I. The parameter P_0 thus represents the intrinsic spin polarization of the ferromagnet measured by PCAR. For small Z, Eq. (5) reproduces the suggested empirical second order polynomial and, therefore, the P_0 values for Co and Fe agree with those reported by Strijkers *et al.*[6]

To demonstrate that the exponential-like decay of P with Z^2 is general and not limited to the case of the transition metals Co and Fe, we consider two other types of materials. Figure 5 shows our results of PCAR measurements on the magnetic rare-earth metal Gd, revealing a decay similar to Co and Fe. The P_0 obtained from a fit of Eq. (5) is 0.45 ± 0.04, which is significantly larger than the tunneling spin polarization of 0.14 ± 0.03 measured with SPT.[2] Ji *et al.*[8] performed PCAR measurements on half-metallic CrO_2 using Pb tips, in an experimental approach equivalent to the one used in this work. They presented P as a function of Z, and used the empirical polynomial expression to extract P_0. Here we plot in Fig. 5 their data as a function of Z^2 and apply Eq. (5).

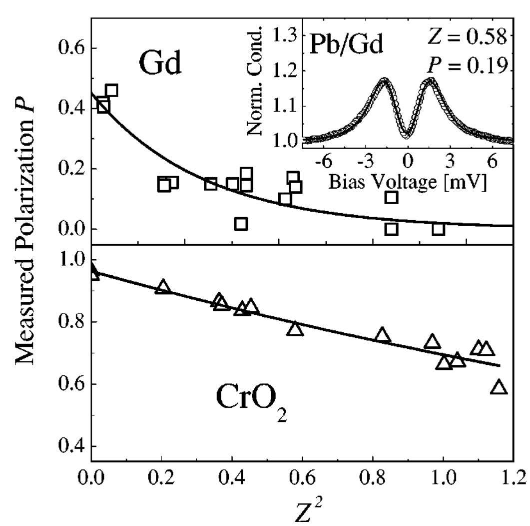

FIG. 5. Measured polarization plotted as a function of Z^2 for Gd and half-metallic CrO_2 (the latter taken from Ref. 8). The solid lines are fits of Eq. (5). The inset shows a representative measurement of a Pb/Gd contact measured at 4.2 K with a BTK fit (solid line).

In comparison with Co, Fe, and Gd, the decay of P with Z^2 is much weaker. The fit results of Gd and CrO_2 are included in Table I.

The coarse experimental approach used presently, does not allow for controlled preparation of the S/N interface. Accordingly, it is very difficult to unambiguously identify and localize scattering contributions to Z^2 and responsible spin-flip mechanisms. Nevertheless, one may speculate on the mechanisms involved based on the experimental observations. The width l in Eq. (3) is related to the characteristic dimension of the region where Andreev reflection occurs. If the bulk of the electrons are Andreev reflected in the proximity region in S, within a superconducting coherence length from the interface, spin-flip scattering occurs due to the spin-orbit interaction[16,17] in the superconductor. However, since the spin-orbit interaction in Pb is at least one order of magnitude larger as compared to Nb, the lack of a dependence on the choice between Pb or Nb as the superconducting tip seems to rule out this mechanism. A more important contribution might be spin flip due to the interaction between the electron magnetic moment and atomic magnetic moments in an atomically intermixed interface region. This picture seems reasonable in view of the contacting procedure used, where short voltage pulses are applied to alter the contact resistance, possibly giving rise to electromigration. The mechanical properties of the sample also have strong influence on the resulting atomic structure of the interface during formation of the contact. The smaller $\alpha\psi$ value for CrO_2, i.e., a weaker decay with Z^2, might be correlated with a significant smaller amount of atomic intermixing at the interface due to the relatively strong bonds in the ionic CrO_2 crystal as compared to the bonding in the other materials.

The considerable spread in P for a given Z^2, which is not caused by limited accuracy of the fits, reflects the importance of the interface controllability. Accordingly, we believe that the measured spin polarization depends to some extent on the delicacy of the applied experimental procedure. Under UHV conditions with clean samples and tips, or with contacts formed by deposition in combination with nanostructuring enabling controlled variation of Z^2, fitted values for $\alpha\psi$ might differ to some degree from those in Table I, and the spread in P may be reduced.

In conclusion, we have performed PCAR measurements on Co and Fe to study the decrease of measured spin polarization with Z. A simple model describing spin-flip scattering in an extended interface region identifies Z^2 as the effective scattering parameter and predicts that the spin polarization decays exponentially with Z^2. In addition to Co and Fe, we have shown the similarity between the model calculations and PCAR data obtained on Gd and half-metallic CrO_2.

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