Spin-Dependent Transparency of Ferromagnet/Superconductor Interfaces

K. Xia,[1] P. J. Kelly,[1] G. E. W. Bauer,[2] and I. Turek[3]
[1]*Faculty of Applied Physics and MESA+ Research Institute, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands*
[2]*Department of NanoScience, Delft University of Technology, 2628 CJ Delft, The Netherlands*
[3]*Institute of Physics of Materials, Academy of Sciences of the Czech Republic, CZ-616 62 Brno, Czech Republic*
(Received 27 February 2002; published 30 September 2002)

We combine parameter-free calculations of the transmission and reflection matrices for clean and dirty interfaces with a scattering-theory formulation of Andreev reflection (AR) generalized to spin-polarized systems in order to critically evaluate the use of an extended Blonder-Tinkham-Klapwijk (BTK) model to extract values of the spin polarization for ferromagnetic metals from measurements of point-contact AR. Excellent agreement with the experimental conductance data is found for Pb/Cu but it is less good for Pb/Ni and poor for Pb/Co, indicating that the BTK formalism does not describe transport through superconducting/ferromagnetic interfaces correctly.

DOI: 10.1103/PhysRevLett.89.166603 PACS numbers: 72.25.Ba, 71.15.Ap, 74.80.Fp

Andreev reflection (AR) spectroscopy at ferromagnet-superconductor (F/S) interfaces in ballistic point contacts (PCAR) has been proposed as a simple method for measuring the spin polarization of ferromagnetic materials [1–3]. When the diameter of the point contact is smaller than the mean-free path but larger than the Fermi wavelength, transport is governed by the Sharvin resistance [4], appropriately modified to take into account the interface "transparency." In the Blonder-Tinkham-Klapwijk (BTK) theory [5], this is done by representing the interface as a planar δ-function barrier between free-electron materials. The Bogoliubov-de Gennes equation [6] for a normal-superconducting (N/S) interface is readily solved for this model and the essential physics expressed in terms of a dimensionless barrier strength Z and the superconducting energy gap Δ. At the interface to a ferromagnetic material, the Andreev reflection is suppressed by the difference between majority and minority electronic band structures [7]. Using the parabolic-band Stoner-Wohlfarth model for ferromagnetic materials, BTK theory can easily be generalized to spin-polarized systems. With only a single additional parameter P, the spin polarization of the ferromagnet, the experimental data can be fitted essentially perfectly [1,3].

This is very surprising because theoretical work in the field of giant magnetoresistance [8] has led to the now generally accepted view that the reflection of conduction electrons at an interface between a nonmagnetic (N) and a ferromagnetic metal (F) is strongly spin dependent. The origin of this spin dependence lies in the different Fermi surfaces and orbital character of the exchange-split d electrons which are responsible for the itinerant ferromagnetism in materials such as Fe, Co, and Ni and of the nonmagnetic metal. Yet the model used to fit the voltage dependent conductance, $G(V)$, measured in PCAR experiments on such ferromagnets, is carried out without introducing a spin-dependent interface transparency parameter Z, and the orbital character of the electrons (for example, in the form of hopping matrix elements across the interface) is completely neglected. In spite of the attractive simplicity of the BTK model, the simple generalization cannot be correct.

In this Letter, we present a generalized scattering formalism of Andreev reflection [9] to spin-polarized systems and calculate $G(V)$ with and without interface disorder. By basing our study on *ab initio* calculations, we can treat the full complexity of the transition metal band structure for specific materials without introducing any arbitrary parameters. We find excellent agreement with experiment for Pb/Cu, somewhat less good agreement for Pb/Ni, and poor results for Pb/Co. It is the poor agreement for the F/S contacts rather than the good agreement for the N/S contact which we want to emphasize. We believe that the generalized BTK model omits an important aspect of the physics of Andreev reflection for ferromagnetic materials which needs to be addressed, and that this point has been obscured by the good fit to experimental data achieved using two-parameter BTK models [10].

We start with the Bogoliubov-de Gennes [6] equation for the electron and hole wave functions of a superconductor:

$$\begin{pmatrix} H_{0\sigma} & \Delta \\ \Delta^* & -H^*_{0-\sigma} \end{pmatrix} \begin{pmatrix} C_{e\sigma} \\ C_{h-\sigma} \end{pmatrix} = \varepsilon \begin{pmatrix} C_{e\sigma} \\ C_{h-\sigma} \end{pmatrix}, \qquad (1)$$

where $\sigma = \pm 1$. $H_{0\sigma}$ is the single-electron Hamiltonian matrix for majority ($\sigma = 1$) and minority ($\sigma = -1$) spins. $C_{e\sigma(h-\sigma)}$ are the coefficient vectors of the wave functions of electrons (holes) in some convenient basis. The excitation energy ε is measured relative to the Fermi energy. Following Beenakker [9], wave-function matching at the F/S interface is achieved by inserting between F and S a fictitious region in which S assumes its normal state (disregarding the proximity effect). At the F/N interface, the scattering of the Bloch states for electrons and holes can be written

$$\begin{pmatrix} C^-_{e\sigma}(F) \\ C^+_{e\sigma}(N) \\ C^+_{h-\sigma}(F) \\ C^-_{h-\sigma}(N) \end{pmatrix} = \begin{pmatrix} S^\sigma(\varepsilon) & 0 \\ 0 & S^{-\sigma*}(-\varepsilon) \end{pmatrix} \begin{pmatrix} C^+_{e\sigma}(F) \\ C^-_{e\sigma}(N) \\ C^-_{h-\sigma}(F) \\ C^+_{h-\sigma}(N) \end{pmatrix}, \quad (2)$$

where + (−) denotes right (left) going waves and

$$S^\sigma(\varepsilon) = \begin{pmatrix} r^\sigma_{11}(\varepsilon) & t^\sigma_{12}(\varepsilon) \\ t^\sigma_{21}(\varepsilon) & r^\sigma_{22}(\varepsilon) \end{pmatrix} \quad (3)$$

is the normal-state scattering matrix. The subscript 1 refers to F, 2 to N. At the (fictitious) N/S interface, there is only Andreev scattering:

$$C^-_{e\sigma}(N) = \alpha C^-_{h-\sigma}(N) e^{i\phi}, \quad (4)$$

$$C^+_{h-\sigma}(N) = \alpha^* C^+_{e\sigma}(N) e^{-i\phi}, \quad (5)$$

where, for $|\varepsilon| < \Delta_0$, $\alpha = \exp[-i \arccos(\varepsilon/\Delta_0)]$ describes the phase shift due to the penetration of the wave function into the superconductor and, for $|\varepsilon| > \Delta_0$, $\alpha = [\varepsilon - \mathrm{sgn}(\varepsilon)\sqrt{\varepsilon^2 - \Delta_0^2}]/\Delta_0$, $|\alpha| < 1$. The incoming and reflected waves are related on the F side by

$$\begin{pmatrix} C^-_{e\sigma}(F) \\ C^+_{h-\sigma}(F) \end{pmatrix} = \begin{pmatrix} R^\sigma_{ee} & R^\sigma_{eh} \\ R^\sigma_{he} & R^\sigma_{hh} \end{pmatrix} \begin{pmatrix} C^+_{e\sigma}(F) \\ C^-_{h-\sigma}(F) \end{pmatrix}. \quad (6)$$

$$R^\sigma_{ee} = r^\sigma_{11}(\varepsilon) + \alpha^2 t^\sigma_{12}(\varepsilon) r^{-\sigma*}_{22}(-\varepsilon) \frac{1}{1 - \alpha^2 r^\sigma_{22}(\varepsilon) r^{-\sigma*}_{22}(-\varepsilon)} t^\sigma_{21}(\varepsilon), \quad (7)$$

$$R^\sigma_{he} = \alpha^* e^{-i\phi} t^{-\sigma*}_{12}(-\varepsilon) \frac{1}{1 - \alpha^2 r^\sigma_{22}(\varepsilon) r^{-\sigma*}_{22}(-\varepsilon)} t^\sigma_{21}(\varepsilon), \quad (8)$$

are the reflection coefficients for the total system. In terms of the spectral conductance

$$G_{FS}(\varepsilon) = \frac{e^2}{h} \sum_{\sigma=\pm 1} \mathrm{Tr}(1 - R^\sigma_{ee} R^{\sigma\dagger}_{ee} + R^\sigma_{he} R^{\sigma\dagger}_{he}), \quad (9)$$

and the Fermi-Dirac distribution function f, the current-voltage relation is $eI(V) = \int d\varepsilon [f(\varepsilon) - f(\varepsilon + eV)] G_{FS}(\varepsilon)$. The charge transport in the presence of Andreev reflection is thus expressed in terms of the scattering matrix for the normal state. This equation is the spin-polarized generalization of the multichannel expression discussed in Ref. [9] and reduces to the BTK equivalent if we use the scattering matrix of a δ-function potential and the parabolic-band model. However, the scattering matrix formulation includes the dependence of the matching of the wave functions at the interface on the symmetry of the bulk states, an effect which is normally neglected even though it can be large [11]. In general, there is no simple dependence on the bulk polarization of the ferromagnet. Note that the expressions also hold for energies above the superconducting gap.

The parameter-free calculation of the reflection matrices [11] and the conductance (9) is based on the surface Green's function method [12] implemented with a tight-binding linear muffin tin orbital basis [13]. Because a minimal basis set is used, we are able to carry out calculations for large lateral supercells and model disorder very flexibly within such supercells without using any adjustable parameters. The electronic structure is determined self-consistently within the local spin density approximation. For disordered layers, the potentials are calculated using the layer coherent potential approximation [12].

We focus our attention here on the Pb/Cu, Pb/Co, and Pb/Ni systems investigated experimentally in [1] which were grown with an fcc (111) orientation in point contacts. We encounter the practical difficulty that the lattice constants of Pb are quite different from those of Cu, Co, and Ni. Lattice matching is modeled using lateral supercells containing 4×4 Cu (Co or Ni) atoms on one side of the interface and 3×3 Pb atoms/monolayer on the other side or 7×7 Cu, Co, or Ni and 5×5 Pb atoms/monolayer, which gives better matching but is computationally more expensive. Disorder is taken into account by introducing a two monolayer-thick 50% interface alloy [11]. The results are not very sensitive to moderate variations in the alloy concentration in this range. Experiments [1] have been carried out at slightly elevated temperatures to suppress a possible proximity effect. Our computations are therefore carried out using the corresponding values of kT in the Fermi function: 4.2 K for Pb/Cu, 1.4 K for Pb/Co, and 2.5 K for Pb/Ni. The only parameter in the calculations is the superconducting energy gap Δ_{Pb} which is taken from experiment and is very close to the bulk value.

For an ideal N/S interface, Andreev reflection leads to a doubling of the conductance. In Table I, the zero bias values $G_{FS}(0)/G_{FN}(0)$, calculated for clean and alloy interfaces, are compared with the experimental values of Ref. [1]. Suppression of the Andreev reflection is apparent in all three cases but is smallest for nonferromagnetic Cu. Even there, however, band structure mismatch between Cu and Pb gives rise to a significant reduction of the interface transparency and suppression of AR. The agreement with experiment is remarkably good, and only in the case of Pb/Co is there any indication of a discrepancy. This picture changes when we compare the calculated and experimental spectra in Fig. 1 shown in terms of $g(V) \equiv [G_{FS}(V) - G_{FN}(0)]/G_{FN}(0)$, where $G_{FN}(0)$ is the normal-state conductance and $G_{FS}(V)$ is the differential

TABLE I. Zero bias suppression of Andreev reflection compared with the experimental values obtained from Ref. [1].

$G_{FS}(0)/G_{FN}(0)$	Pb/Cu	Pb/Ni	Pb/Co
Clean interface	1.54	1.29	1.08
Alloy interface	1.36	1.16	1.00
Experiment	1.38	1.18	1.13

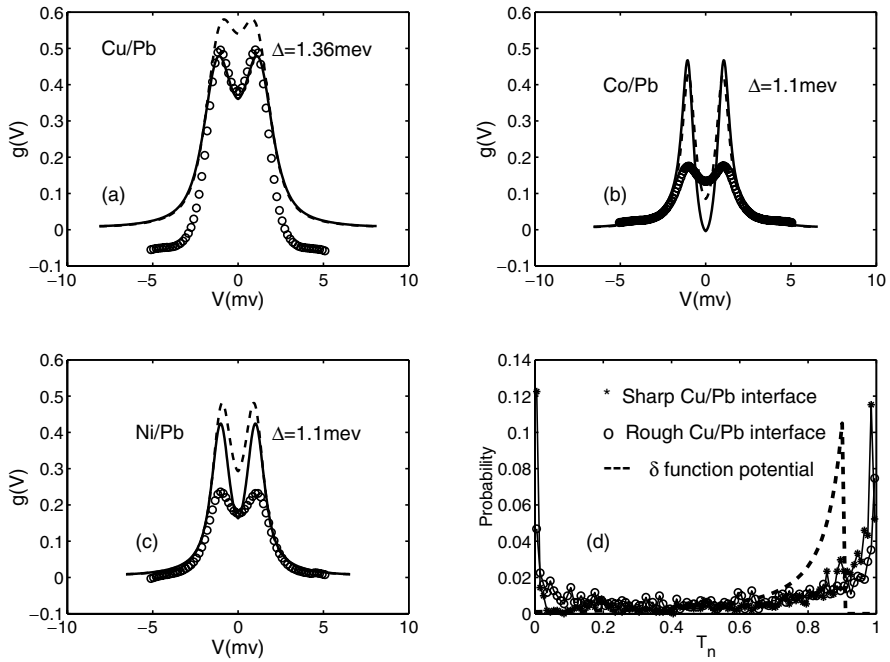

FIG. 1. (a)–(c) Calculated $g(V)$ curves for disordered (solid line) and clean interfaces (dashed line) compared with the Cornell group's experimental results (o). $g(V) \equiv [G_{FS}(V) - G_{FN}(0)]/G_{FN}(0)$, where $G_{FN}(0)$ is the normal state conductance and $G_{FS}(V)$ is the differential conductance for an F/S interface at finite temperature and finite bias. (a) Pb-Cu, (b) Pb-Co, and (c) Pb-Ni. (d) Distribution function, $\rho(T)$, of the transmission matrix eigenvalues of a clean (x) and disordered (o) Pb/Cu interface. The distribution function of the BTK model with $Z = 0.3$ is plotted as a dashed line for comparison.

conductance for an F/S interface at finite temperature and finite bias.

Let us first focus on the paramagnetic Pb/Cu point contact, Fig. 1(a). Whereas results for a specular interface deviate significantly from the measured data, the decreased conductance of a rough interface leads to essentially perfect agreement with experiment. Aspects of the problem not taken into account, such as spin-orbit scattering and strong-coupling superconductivity in Pb are apparently not very important.

The agreement with the PCAR spectra for the ferromagnetic systems [Figs. 1(b) and 1(c)] is not satisfactory close to the band edge of quasiparticle excitations, where the theoretical results appear to strongly exaggerate the Andreev reflection [14]. Since we have no parameters with which we could improve the agreement, the origin of the deviations should be sought in deficiencies of the underlying model. Whereas a proximity effect can be excluded for single point contacts whose diameters are smaller than the superconducting coherence length, the Zeeman splitting of the superconducting density of states due to stray fields from the ferromagnet may account for the discrepancies [15,16].

The spin polarization of the interface conductance, $P = (G_{\mathrm{maj}} - G_{\mathrm{min}})/(G_{\mathrm{maj}} + G_{\mathrm{min}})$, will, in general, differ from that of the bulk ferromagnetic material. For example, for Pb/Ni, $P = 0.5\%$ for a specular interface and -1.4% for a rough interface. This should be compared to the BTK fit obtained in Ref. [1] of $P_{\mathrm{BTK}} = 32\%$. Our prediction of a small spin dependence of the interface conductance could be tested directly by measuring the magnetoresistance of a Ni/Pb/Ni trilayer (or multilayer) in the current-perpendicular-to-the-plane configuration with the layer thickness chosen sufficiently thin that the total resistance is dominated by the interface contribution. Using the expression given by Schep *et al.* [17] for diffuse systems, we predict interface resistances of $R_{\mathrm{maj}} = 0.97\, f\Omega m^2$ and $R_{\mathrm{min}} = 1.27\, f\Omega m^2$ for a rough Pb/Ni (111) interface.

To emphasize the difference between the single-channel BTK model and the multichannel scattering-theory generalization, we focus on the zero-voltage, zero-temperature conductance (10) for a nonmagnetic system:

$$G_{NS} = \frac{4e^2}{h} \sum_n \frac{T_n^2}{(2 - T_n)^2} = \frac{4e^2}{h} \int dT \rho(T) \frac{T^2}{(2 - T)^2} \quad (10)$$

is expressed in terms of the eigenvalues of the transmission matrix, T_n, and the sum is over the propagating channels [9]. For the Pb/Cu interface, the distribution function of transmission matrix eigenvalues, $\rho(T)$, is shown in Fig. 1(d). It turns out to be a bimodal function with a clear dependence on interface roughness and on the constituent materials. Obviously, $\rho(T)$ strongly affects the Andreev reflection probability with the main contribution coming from the highly transmitting part, whereas the average conductance is much more affected by the low transmission channels. Using the Andreev conductance to derive an average transmission is therefore misleading.

For a δ-function potential and free electrons, the transmission probability as a function of the in-plane wave vector has the form $T_{k_\parallel} = 1/\{1 + Z^2/[1 - (k_\parallel/k_F)^2]\}$. The distribution function for $Z = 0.3$, the value which must be taken to reproduce the average conductance, is plotted as a dashed curve in Fig. 1(d) and is seen to be qualitatively incorrect compared to the realistic distributions.

For the F/S interface, the situation is more complicated. Since Cooper pairs are phase sensitive, the conductance is governed by the full scattering matrix, i.e., by the scattering amplitudes and not just the transmission probabilities. We therefore cannot use a single distribution function to characterize the interface because the Andreev reflection probability contains terms such as $1 - \alpha^2 r_{22}^{\sigma}(\varepsilon) r_{22}^{-\sigma *}(-\varepsilon)$ which cannot be expressed by $T_\uparrow$ and $T_\downarrow$. Still, it should be clear that the deficiencies of the δ function barrier potential will not vanish.

In summary, we have calculated the PCAR spectra of Pb/Co, Pb/Ni, and Pb/Cu point contacts and the spin polarization of the interface conductances from first principles which allows us to remove a number of dubious approximations made when applying the BTK model to transition metals. Though we find good agreement with experimental results by the Cornell group for the full spectrum of Pb/Cu and for the zero bias conductance in the ferromagnetic systems, qualitative discrepancies are revealed for the full spectra of the ferromagnetic materials indicating some interaction between the ferromagnet and superconductor not included in the BTK model such as the Zeeman splitting of the superconducting densities of states caused by the stray magnetic fields of the ferromagnet. The good fitting achieved with two-parameter free-electron BTK models has obscured the need to consider such interactions. The scattering theoretical expression for the conductance indicates that PCAR measures a spin-dependent interface transparency and is not suitable for determining the polarization characterizing bulk ferromagnets.

Our results have more general relevance in the field of mesoscopic transport studies. There is a general misconception that a sufficiently clean (defect-free) interface between two metals will result in perfect transparency or, in the worst case, in a transparency which is reduced by the mismatch of Fermi velocities. We note that mismatch of Fermi velocities can be very large for transition metals with complex Fermi surfaces. In addition, however, the mismatch of the wave-function character at the Fermi energy can, and does, give rise to reduced transparency which is equally important. The discrepancy between simple model studies of PCAR and our calculations provides ample evidence that it is very important to take these factors into account when interpreting the results of transport studies in inhomogeneous materials.

This work is supported by the Stichting FOM, the NEDO International Joint Research Grant Program "Nano-magnetoelectronics," the Grant Agency of the Czech Republic (202/00/0122), the European Commission's TMR Research Network on "Interface Magnetism" (Contract No. FMRX-CT96-0089) and RT Network "Computational Magnetoelectronics" (Contract No. HPRN-CT-2000-00143). We acknowledge fruitful discussions with W. Belzig, A. Brataas, Yu. Nazarov, and J. Kudrnovsky and thank R. A. Buhrman for providing us with the experimental data shown in Fig. 1.

[1] S. K. Upadhyay *et al.*, Phys. Rev. Lett. **81**, 3247 (1998); Appl. Phys. Lett. **74**, 3881 (1999).
[2] R. J. Soulen *et al.*, Science **282**, 85 (1998); B. Nadgorny *et al.*, Phys. Rev. B **61**, R3788 (2000).
[3] Y. Ji *et al.*, Phys. Rev. Lett. **86**, 5585 (2001).
[4] Y.V. Sharvin, Zh. Eksp. Teor. Fiz. **48**, 984 (1965) [Sov. Phys. JETP **21**, 655 (1965)].
[5] G. E. Blonder, M. Tinkham, and T. M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).
[6] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Addison-Wesley, Reading, MA, 1966).
[7] M. J. M. de Jong and C. W. J. Beenakker, Phys. Rev. Lett. **74**, 1657 (1995).
[8] For a review, see M. A. M. Gijs and G. E. W. Bauer, Adv. Phys. **46**, 285 (1997).
[9] C. W. J. Beenakker, Rev. Mod. Phys. **69**, 731 (1997); C. J. Lambert and R. Raimondi, J. Phys. Condens. Matter **10**, 901 (1998), and references therein.
[10] This point has been made by a number of authors using various extensions of BTK theory. See I. I. Mazin, Phys. Rev. Lett. **83**, 1427 (1999); I. I. Mazin, A. A. Golubov, and B. Nadgorny, J. Appl. Phys. **89**, 7576 (2001); I. Zutic and O. T. Valls, Phys. Rev. B **60**, 6320 (1999); Phys. Rev. B **61**, 1555 (2000). Unlike the present work, however, such parametrized models do not allow comparison to be made to specific materials preventing definite conclusions being drawn.
[11] K. Xia *et al.*, Phys. Rev. B **63**, 064407 (2001).
[12] I. Turek *et al.*, *Electronic Structure of Disordered Alloys, Surfaces and Interfaces* (Kluwer, Dordrecht, 1997).
[13] O. K. Andersen, O. Jepsen, and D. Glötzel, in *Highlights in Condensed Matter Theory*, edited by F. Bassani, F. Fumi, and M. P. Tosi (North-Holland, Amsterdam, 1985), p. 59.
[14] Similar results for clean Pb/Cu and Pb/Co interfaces have been reported by Taddei, Sanvito, and Lambert [J. Low Temp. Phys. **124**, 305 (2001)] using a multiband tight-binding model. In our self-consistent calculations for Pb/Co, we find no evidence for an enhancement of the exchange splitting at the interface which these authors invoke to obtain agreement with experiment for Pb/Co.
[15] W. Belzig (private communication).
[16] R. Mélin, Europhys. Lett. **51**, 202 (2000).
[17] K. M. Schep *et al.*, Phys. Rev. B **56**, 10 805 (1997).