Abstract. We examine here electronic transport in nanoscale systems where normal and ferromagnetic probes are attached to a conventional superconductor. While reviewing the long-studied effects of Andreev reflection and charge imbalance, we concentrate on two recently predicted coherent, nonlocal processes known as crossed Andreev reflection and elastic co-tunnelling. These processes can occur when two spatially separated normal or ferromagnetic probes are separated by a distance comparable to the coherence length of the superconductor. Here we show that normal probes, by avoiding some of the experimental and theoretical complications of ferromagnetic probes, may offer a better opportunity to examine these processes.
1. Introduction

Although the physics of a normal metal (N) in contact with a superconductor (S) has been studied extensively for many years, the subject has seen renewed interest in recent years with the observation of novel effects in micron size normal-metal/superconductor (NS) heterostructures [1]. In the case of a clean normal metal in contact with a superconductor through a barrier of arbitrary strength, the conductance of the interface can be described well by the model of Blonder, Tinkham and Klapwijk (BTK) [2], which predicts, for example, a factor of two increase in the conductance of a highly transparent NS interface at low temperatures. For a dirty normal metal (the situation of interest here), where the motion of electrons is diffusive, the situation is more complicated, and one needs to take recourse to the quasiclassical theory of superconductivity (see, for example [3]). In contrast to clean systems, the characteristic energy scale for diffusive systems is not set by the energy gap $\Delta$ of the superconductor, but by the Thouless energy $E_c = \hbar D / L^2$, where $D = (1/3) v_F \ell$ is the electronic diffusion coefficient, $v_F$ the Fermi velocity, $\ell$ the elastic mean free path, and $L$ the relevant length of the sample. With modern lithographic capabilities, this now corresponds to an experimentally accessible energy scale, and many experiments have been performed to look at new aspects of the superconducting proximity effect [4]–[13].

The microscopic basis of the proximity effect is the process of Andreev reflection [14], whereby an electron incident on a NS interface from the normal metal with an energy $E$ above the Fermi energy $E_F$ combines with another electron with an energy $-E$ below $E_F$ to form a Cooper pair that propagates into the superconductor. The second electron that is removed from the Fermi sea can be thought of as a hole that propagates away from the interface; Andreev reflection is therefore the reflection of an incident electron as a hole. If the electron phase coherence length $L_\phi$ in the metal is long enough, the electron and its retroreflected hole are phase-coherent. Many experiments have elegantly demonstrated this phase coherence in the electrical and thermal transport of NS structures.
More recently, interest has arisen in structures in which the superconductor is in contact with a ferromagnet (F). For conventional Andreev reflection into a \( s \)-wave superconductor, the two electrons that combine must have opposite spin orientations. In an itinerant ferromagnet in which the conduction electron cloud is spin polarized, not all the electrons which are incident on the FS interface can find complementary electrons of the opposite spin polarity to form a Cooper pair, resulting in a decrease in the probability of Andreev reflection that is directly related to the degree of spin polarization in the ferromagnet. This leads to a corresponding decrease in the conductance of the FS junction in comparison to an equivalent NS junction [16]–[20]. The decrease in the conductance of the FS junction has been used effectively to measure the degree of spin polarization in the ferromagnet F [21]–[23].

Interest has also focused on the possibility of observing long-range superconducting correlations in ferromagnets [24, 25], in spite of the presence of an exchange field which might normally be expected to destroy any superconducting correlations. For the conventional spin-singlet case, the superconducting correlations decay over the very short length scale of the exchange length \( \xi_E = \hbar v_F/k_B T_{\text{Curie}} \) (or \( \xi_E = \sqrt{\hbar D/k_B T_{\text{Curie}}} \) in a diffusive ferromagnet); here \( T_{\text{Curie}} \) is the Curie temperature. The mismatch of the wavevectors for the spin-up and spin-down electrons also leads to oscillations of the superconducting order parameter in the ferromagnet, leading to the possibility of \( \pi \) phase coupling in SFS junctions [15], [26]–[28]. It has also been theoretically predicted that it should be possible to nucleate spin-triplet superconducting correlations in a ferromagnet in contact with a superconductor that decay over the much longer length scale of the usual normal-metal superconducting coherence length \( \xi_N = \hbar v_F/k_B T \) (or \( \xi_N = \sqrt{\hbar D/k_B T} \) in the diffusive case). Very recently, there have been reports of such spin-triplet superconductivity in the half-metallic ferromagnet CrO\(_2\) [29] and the conical ferromagnet Ho [30].

The majority of the work on NS and FS structures to date has focused on superconducting correlations in a single normal metal or ferromagnet that is in contact with a superconductor. However, recent theoretical work has highlighted the possibility of observing coherent correlations induced between quasi-particles in spatially separated normal-metals and ferromagnets, coupled through their mutual interaction with a superconductor [31]. These processes are essentially non-local versions of the Andreev reflection process, and are called crossed Andreev reflection (CAR) and elastic co-tunnelling (EC). Intricately tied to this general area is quasiparticle injection from normal metals into superconductors, a topic that had been investigated extensively three decades ago, but which has not been discussed theoretically in connection with nonlocal effects. In this paper, we discuss our experiments on local and nonlocal transport in FS and NS devices, concentrating in particular on experimental results that remain to be understood. While the data for the FS devices is presented here for the first time, much the NS experimental work has been reported previously [32].

2. CAR, EC and charge imbalance

2.1. CAR and EC

Figure 1(a) shows a schematic representation of conventional Andreev reflection: a spin-up electron and energy \( E \) incident on a NS interface can propagate into the superconductor as a

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1 An excellent review of proximity effects in superconductor-ferromagnet devices can be found in the review article [15].
Cooper pair by coupling with a second spin-down electron of energy \(-E\) in the same normal metal. The second electron that is removed from the Fermi sea can be thought of as a hole that travels away from the NS interface. Clearly if the number of spin-up electrons is greater than the number of spin-down electrons, as would be in the case of a spin-polarized ferromagnet, the probability of spin-up electrons pairing up with spin-down electrons to form Cooper pairs is lower, and hence the probability of Andreev reflection is reduced in a FS interface.

The two electrons that couple to form the Cooper pair need not come from the same normal metal. Figure 1(b) shows the case of CAR, where the spin-up electron comes from one normal metal (say the left normal metal, labelled 1 in the figure) and the spin-down electron comes from a second normal metal (the right normal metal, labelled 2 in the figure). In order for this to occur, the distance between the two normal metals in the superconductor must be comparable to the superconducting coherence length \(\xi_S\). Since the electrons in the two normal metals are coupled via their interaction with the superconductor, if one drives a current from normal metal 1 into the superconductor through the NS interface, a current will also be induced from normal metal 2 into the superconductor. Experimentally, it is easier to measure a voltage rather than a current;
if one measures the voltage on normal metal 2 with respect to the superconductor in response to the drive current under the condition that no current flows across the second NS interface, then the voltage should be negative for CAR. This voltage is nonlocal in the sense that the voltage is measured in a region where there is no drive current.

The spin-up electron from normal metal 1 can also tunnel through the superconductor directly into normal metal 2, in an effect termed EC. In this case, the spin is preserved, and as in CAR, one can induce a current through the second interface in response to a drive current through the first NS interface. Figure 1(c) shows that the sign of the induced current is opposite that in CAR, and hence the sign of the induced voltage is also opposite, i.e. in EC, the sign of the induced nonlocal voltage is positive with the direction of the drive current. In general, CAR and EC are both present, hence the total nonlocal voltage is the sum of the CAR and EC contributions.

The dependence of EC and CAR on the spins of the two electrons has direct implications for the case when the two normal metals are ferromagnets. Consider first the case when the normal metals are half-metallic ferromagnets, so that the electrons in the ferromagnet are completely spin-polarized. If the magnetizations of the two ferromagnets are aligned, then the spin-up electron in ferromagnet 1 cannot find a corresponding spin-down electron in ferromagnet 2 to form a Cooper pair in the superconductor. Hence the process of CAR is completely suppressed in this case. However, a spin-up electron from the first ferromagnet can tunnel directly into the second ferromagnet since there are allowed spin-up states in ferromagnet 2, so EC is still allowed. When the magnetizations of the two ferromagnets are anti-aligned, CAR is favoured and EC completely suppressed. Hence, investigating a FSF system allows one in principle to distinguish between CAR and EC. In a real experiment, however, one has ferromagnets with finite spin-polarizations $P$, so that it is more difficult to separate the CAR and EC contributions.

If we consider the three terminal geometry shown in figures 1(b) and (c), we can represent the currents $I_1$ and $I_2$ going through the two NS interfaces in terms of the voltages $V_1$ and $V_2$ on each normal electrode with respect to the superconductor [33]

$$
\begin{pmatrix}
I_1 \\
I_2
\end{pmatrix} = 
\begin{pmatrix}
G_{A1} + G_{CA} + G_{EC} & G_{CA} - G_{EC} \\
G_{CA} - G_{EC} & G_{A2} + G_{CA} + G_{EC}
\end{pmatrix}
\begin{pmatrix}
V_1 \\
V_2
\end{pmatrix}.
$$

(1)

Here $G_{A1(2)}$ is the conventional Andreev conductance of the first (second) NS interface, $G_{CA}$ is the crossed Andreev conductance and $G_{EC}$ is the EC conductance. As we have noted, it is easier for us to measure nonlocal voltages instead of currents. Inverting equation (1) to write the voltages $V_1$, $V_2$ in terms of the currents, $I_1$, $I_2$, we have

$$
\begin{pmatrix}
V_1 \\
V_2
\end{pmatrix} = \frac{1}{\gamma} 
\begin{pmatrix}
G_{A2} + G_{CA} + G_{EC} & G_{EC} - G_{CA} \\
G_{EC} - G_{CA} & G_{A1} + G_{CA} + G_{EC}
\end{pmatrix}
\begin{pmatrix}
I_1 \\
I_2
\end{pmatrix}.
$$

(2)

where $\gamma = (G_{A1} + G_{CA} + G_{EC})(G_{A2} + G_{CA} + G_{EC}) - (G_{CA} - G_{EC})$ [2]. Thus, if $I_1 = I$ (the drive current) and $I_2 = 0$, then the nonlocal voltage $V_2 = (G_{EC} - G_{CA})I/\gamma$, or the nonlocal resistance $R_{nl} = V_2/I = (G_{EC} - G_{CA})/\gamma$, where $G_{CA}$ and $G_{EC}$ are both positive quantities. From this, we can see that the nonlocal resistance is positive if $G_{EC} > G_{CA}$ and negative if $G_{EC} < G_{CA}$.

The first discussion of CAR and EC by Byers and Flatte [31] examined the conductances for two, tunnelling point contacts on a superconducting surface. Work has since been done developing the theory to cover extended contacts [33], transparent interfaces [34] and diffusive...
Figure 2. Schematic representation of local charge imbalance. The chemical potential in the normal metal $\mu_N$, measured with a normal probe, shows a linear dependence on distance. In the superconductor, the chemical potential of the Cooper pairs $\mu_{cp}$, which relaxes to the bulk chemical potential on a length scale of $\xi_S$, is measured with a superconducting contact, while the chemical potential of the quasiparticles $\mu_{qp}$, which relaxes on the longer length scale $\Lambda_{Q^*}$, is measured with a normal metal contact.

superconductors [35]. In general the conductances will have a spatial dependence of $e^{-a(r/\xi_S)}$ where $a$ is a constant on the order of unity and $r$ the distance between the two normal or ferromagnetic contacts. This spatial dependence is multiplied by an additional geometric factor that varies from $1/r^2$ for single channel tunnel contacts on a ballistic superconductor to 1 for extended contacts on a diffusive superconductor (see Feinberg [35] for an overview). As mentioned above, the relative magnitudes of $G_{EC}$ and $G_{CA}$ when ferromagnetic contacts are used will depend on the polarization of the contacts. For normal metal contacts, the earlier theoretical work predicted that $G_{EC} = G_{CA}$ for tunneling contacts, while more recent studies have shown that $G_{EC} > G_{CA}$ in the clean contact limit [36], an inequality that may also hold in the tunneling regime if the proximity effect is taken into account [37].

2.2. Charge imbalance

In addition to CAR and EC, a third effect may also give rise to a nonlocal resistance in the geometry of figures 1(b) and (c). This is the phenomena of charge imbalance, which is illustrated schematically in figure 2 for the local case, which was studied extensively three decades ago [38]–[40]. Charge imbalance is a nonequilibrium phenomenon that arises from the conversion of quasiparticle current in the normal metal to supercurrent in the superconductor. This conversion gives rise to nonequilibrium chemical potentials for the quasiparticles ($\mu_{qp}$) and the Cooper pairs ($\mu_{cp}$) in the vicinity of the NS interface. $\mu_{cp}$ relaxes to the value of the chemical potential in the bulk of the superconductor over a length scale of $\xi_S$, which is typically in the order of 100 nm for Al in the dirty limit, the superconductor of relevance for the experiments reported here. $\mu_{qp}$ is relaxes over the distance of the charge imbalance length $\Lambda_{Q^*}$. In diffusive metals, which
are the materials of interest here, the charge imbalance length is given by \( \Lambda_{Q^*} = \sqrt{D \tau_{Q^*}} \), where \( D = \frac{1}{3} v_F \ell \) is the diffusion coefficient, \( \ell \) is the elastic mean free path and the charge imbalance time \( \tau_{Q^*} \) close to the superconducting transition has been modelled by [40]

\[
\tau_{Q^*} = \frac{4 k_B T}{\pi \Delta(T)} \tau_{\text{in}}.
\]

Here \( \Delta(T) \) is the temperature dependent gap in the superconductor and \( \tau_{\text{in}} \) is the electron inelastic scattering time. As \( T \to T_c \), \( \Delta(T) \to 0 \) and \( \tau_{Q^*} \) and \( \Lambda_{Q^*} \) diverge. Values of \( \Lambda_{Q^*} \) in the order of tens of microns have been measured in early experiments on phase slip centres very close to the superconducting transition in Al [41]–[43]. Thus \( \Lambda_{Q^*} \) is typically larger than \( \xi_S \) in the experiments performed to date.

The spatial dependence of \( \mu_{qp} \) near the NS interface is given by [44]

\[
\mu_{qp} = e \Lambda_{Q^*} \rho_{\text{Al}} I \tanh(x/\Lambda_{Q^*}),
\]

where \( x \) is the distance from the NS interface into the superconductor and \( \rho_{\text{Al}} \) is the resistance per unit length of the Al wire. This is the chemical potential measured by a normal metal probe. If a superconducting probe were placed at the same position \( x \), it would measure \( \mu_{cp} \), which for distances \( x > \xi_S \) is essentially the chemical potential in the bulk of the superconductor. The bulk value of the chemical potential can be determined by taking the asymptotic value of \( \mu_{qp} \) for \( x \gg \Lambda_{Q^*} \), which is simply \( e \Lambda_{Q^*} \rho_{\text{Al}} I \). Thus, the voltage difference measured between a superconducting probe and a normal probe placed a distance \( x \) from the interface is just \( \Delta V = \Lambda_{Q^*} \rho_{\text{Al}} I [1 - \tanh(x/\Lambda_{Q^*})] \), giving rise to a resistance

\[
\Delta R(x) = \Lambda_{Q^*} \rho_{\text{Al}} [1 - \tanh(x/\Lambda_{Q^*})].
\]

It should be emphasized that since \( \mu_{qp} \) reaches the value in the bulk of the superconductor for \( x > \xi_S \), the resistance measured between the normal voltage probe at \( x \) and any superconducting probe at a distance \( x > \xi_S \) would measure the same \( \Delta R(x) \).

When the superconductor is normal, a voltage probe made from a superconducting material placed at a distance \( x > \xi_S \) from the interface would measure a linearly varying potential \( \mu_N(x) = e \rho_{\text{Al}} I x \), while the same probe below the superconducting transition would measure the bulk chemical superconducting potential that we have just shown is \( e \rho_{\text{Al}} I \Lambda_{Q^*} \). Thus, a superconducting probe would measure an enhancement of the resistance of the NS interface over the normal state resistance \( R_N \) by an amount

\[
\Delta R_{\text{int}}(x) = (\Lambda_{Q^*} - x) \rho_{\text{Al}}.
\]

Since \( \Lambda_{Q^*} \) diverges at the superconducting transition, this resistance enhancement should be maximum at the transition, and decay as the temperature is decreased below \( T_c \). Enhancements of the normal state resistance near the superconducting transition have been seen in a number of experiments on NS interfaces and phase slip centres [45, 46]. However, the resistance enhancement observed is typically in the order of 0.2–0.4 \( R_N \), where \( R_N \) is the normal state resistance of the interface. This should be compared to the resistance enhancements of 5–20 \( R_N \) expected if \( \Lambda_{Q^*} \) is of the order of 10–40 microns as reported in earlier experiments [41]–[43]. If equation (6) is correct, our estimates of \( \Lambda_{Q^*} \) based on these recent experiments must be revised to values of the order of \( \simeq 1 \mu m \) in Al.

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It must be emphasized that the foregoing description of charge imbalance in the electrical transport of NS interfaces is for the local case. To our knowledge, there is no theoretical description yet of charge imbalance in the nonlocal case, by which we mean a geometry similar to that of figures 1(b) and (c), where no current flows through the interface through which the potential is being measured by a normal metal probe. However, it is reasonable to assume that if a charge imbalance is created at the first NS interface through which a current is being driven, it will decay over some length scale in all directions into the superconductor. The length scale over which it decays may be different depending on whether a current is present, so that the effective charge imbalance length may be different in the nonlocal case. In addition, the charge imbalance length would conceivably be modified if the quasiparticle current injected into the superconductor were spin polarized (in the case when the normal metal is a ferromagnet), since the relaxation time would also involve the polarization of the quasiparticles in the superconductor. In either case, the nonlocal voltage measured on the second normal metal probe with respect to the superconductor would be positive, i.e. of the same sign as the contribution due EC, and opposite that due to CAR. The decay length of charge imbalance and EC/CAR as a function of distance between the two interfaces is expected to be different: while EC and CAR contributions decay exponentially on the length scale of $\xi_S$, the decay length of the charge imbalance contribution based on our knowledge of the local case might be expected to decay on a length scale comparable to $\Lambda_{Q^*}$, with a spatial dependence given by equation (5).

In summary, we note here again for reference the sign of the three potential contributions to the nonlocal signal in FSF and NSN structures: CAR, EC and charge imbalance. With reference to the sample geometry of figure 1(b) or (c), with a drive current being driven from the first normal metal and the superconductor and the nonlocal voltage being measured between the second normal metal and the same superconductor, EC and charge imbalance would give rise to a positive nonlocal voltage, while CAR would give rise to a negative nonlocal voltage. The different signs of the effects, coupled with their different spatial dependences give us in principle the potential to distinguish experimentally between them.

3. Experimental techniques: fabrication and measurement

3.1. Fabrication

All the devices discussed in this paper were patterned using conventional electron beam lithography using a JEOL 840 scanning electron microscope converted for e-beam writing. The substrates were oxidized Si wafers. Figure 3 shows an example of a ferromagnet/normal-metal/superconductor device. Since all the devices consist of at least two to three different materials, multilevel patterning was essential. Our electron-beam lithography software allows us to align features on different lithography levels on the devices to better than 20 nm, and in some cases better than 10 nm. For devices incorporating ferromagnetic materials, the ferromagnet was the first layer of the device to be patterned. This was to ensure that the ferromagnetic material lay flat on the substrate surface; if the ferromagnet were to be placed on either the normal metal or superconducting elements of the device, the resulting uneven film profile would be more likely to nucleate ferromagnetic domain structure. For the same reason, the ferromagnetic materials were patterned in the form of ellipses, as our previous studies [47] indicated that this shape profile was likely to orient the magnetization preferentially along the major axis of the ellipse.
Figure 3. False colour scanning electron micrograph of a FNS device fabricated by electron-beam lithography. The ferromagnetic element is patterned in the shape of an ellipse to preferentially align the magnetization along the major axis of the ellipse. The external magnetic field is also aligned along the major axis of the ellipse in the plane of the substrate. The numbers refer to the contacts used in the measurement (see text).

and the magnetization could be controlled easily by an external magnetic field aligned in the same direction. The electrical contacts on the ferromagnetic particles (see figure 3) enabled us to investigate the magnetic behaviour by measuring the particle’s anisotropic magnetoresistance (AMR) [47]. The ferromagnetic materials used were either polycrystalline Ni or permalloy (Ni₈₁Fe₁₉) deposited using an e-gun evaporator, giving resistivities typically around 6 µΩ cm.

The next layer to be patterned and deposited (or the first layer in the case of the NS devices) was the normal metal, which for the devices discussed in this paper was Au. The final deposited layer was the superconductor Al. The resistivities of the Au and Al films were in the range of 2–4 µΩ cm. In order to ensure transparent interfaces between each metallic layer, an in situ Ar⁺ ion plasma etch was performed immediately prior to deposition. This was especially critical for the ferromagnet–superconductor interfaces, as the Ni or permalloy developed a native oxide layer under ambient conditions. With this in situ etch, the specific interface resistance for the Ni/Al interfaces was 20–40 mΩ µm² and the specific resistance for the Al/Au interfaces was 1.4–2.6 mΩ µm², essentially the Sharvin resistance of the interface. For comparison, an external Ar⁺ ion etch, in which the devices were etched in a separate chamber and immediately transferred by hand through air to the evaporator chamber for deposition, gave specific interface resistances of 370 mΩ µm² to 19 Ω µm² for the Ni/Al interfaces, the large range of values reflecting the irreproducible nature of the process.

3.2. Measurement

The devices were measured in a ⁴He refrigerator with a base temperature of 260 mK or a dilution refrigerator with a base temperature of ~16 mK. The ⁴He refrigerator could be inserted in a cryogenic dewar equipped with a two-axis magnet which allowed the application of magnetic
fields both in the plane of the sample substrate and perpendicular to the sample substrate. Since the application of a magnetic field along the axis of the elliptical ferromagnets was essential, all the devices incorporating ferromagnetic elements were measured using the $^3$He refrigerator. The cryostat of the dilution refrigerator was equipped with a large axial superconducting magnet, although only small magnetic fields were required for these experiments. Both refrigerators were well-shielded against interference from rf and line frequency sources.

Local and non-local four terminal resistance measurements were performed using homemade, ac Adler–Jackson type resistance bridges operating at frequencies in the range of 11–200 Hz. By using multiple bridges operating at different frequencies, we were in principle able to simultaneously measure the resistances of multiple sections of a single device. In practice, equipment space considerations limited us to two simultaneous resistance measurements. Ac excitation currents were typically in the range of 20–200 nA in order to avoid self-heating. For the local and nonlocal four-terminal differential resistance measurements, a dc current was applied between two of the contacts on the device using a home-made current source with an output impedance greater than $10^{12} \Omega$. The remaining part of the measurement circuit could be kept the same, enabling us to measure the simultaneous four-terminal differential resistance $dV/dI$ as a function of dc current $I_{dc}$ for two different sections of a single device. Given the number of contacts, we shall adopt the now standard notation for four-terminal measurements: $R_{1,2;3,4}$ denotes the four-terminal resistance where the current is introduced through contact 1 ($I^+$) and removed through contact 2 ($I^-$) and the voltage is measured across contacts 3 and 4 ($V^+$ and $V^-$ respectively). For the differential resistance, we shall adopt the same notation, i.e. $R_{1,2;3,4}$ denotes $dV_{3,4}/dI_{1,2}$, where it will be clear from the context whether we are referring to a zero-bias resistance or a differential resistance as a function of dc current. For the differential resistance measurements, it is necessary to also give the path of the dc current $I_{dc}$; this will be given explicitly for each case.

4. Experimental results

4.1. Ferromagnet/superconductor/normal-metal structures

4.1.1. Structures with a single ferromagnet. Figure 4(a) shows the temperature dependence of the zero-bias resistances $R_{1,6;10,9}$ and $R_{1,6;10,3}$ of a FS interface in a device similar to that shown in figure 3. For both four-terminal configurations, the ac current is sent from the Ni particle to the superconductor, and the voltage is measured using a common normal contact on the ferromagnet as $V^+$ (lead 10) and a contact on the superconductor as $V^-$. For the red curve, the $V^-$ contact is Al (lead 9), while for the blue curve, the $V^-$ contact is Au. Above $T_c \sim 1.2$ K of the superconductor, both resistances show no temperature dependence and are essentially the same. As the temperature is decreased, the red curve which is measured with a superconducting $V^-$ contact shows a large peak in the resistance at $T_c$. This is the signature of charge imbalance discussed above, and only appears if a superconducting contact is used as a voltage probe: the blue curve, measured with a normal $V^-$ contact shows no resistance peak. As the temperature is decreased further, the peak in the red curve decays, and it approaches the blue curve. However, both curves still show a temperature dependence at the lowest temperatures which is associated with Andreev reflection at the FS interface [49].

2 A detailed description of our typical experimental measurement techniques can be found in [48].
In order to eliminate the temperature dependence of the interface, one can subtract the blue curve from the red curve, which corresponds to measuring the four-terminal resistance $R_{1,6;3,9}$. Since the superconducting contact (lead 9) and the normal-metal contact (lead 3) are effectively measuring the potential at the same distance from the FS interface, classically one might expect the measured resistance to vanish. As can be seen in figure 4(b), this is essentially what occurs above $T_c$; the difference in resistance is zero. Below $T_c$, however, a large peak appears. Comparison to the schematic in figure 2 shows that what is being measured is the difference between $\mu_{\text{qp}}$ and $\mu_{\text{cp}}$ at the same point, i.e. the peak in figure 4(b) is a pure charge imbalance effect. As the temperature is decreased further, the charge imbalance contribution decreases and appears to vanish, although there is some indication of a small but finite resistance even at the lowest temperatures, a potential contribution from CAR and EC. We note that the magnitude of the resistance enhancement is $\sim 1.65 \Omega$. From equation (6), the enhancement of the resistance over the normal state resistance is $\rho_{\text{Al}}(\Lambda_{Q^*} - x)$. For this sample, $\rho_{\text{Al}} = 5.6 \Omega/\mu$m$^{-1}$ and $x = 110$ nm, giving $\Lambda_{Q^*} = 0.41 \mu$m, far smaller than the values found in the early studies on charge imbalance in Al.
A similar dependence can be observed in the differential resistance $dV/dI$ as function of $I_{dc}$. Figure 5(a) shows differential resistances $R_{1,6;3,9}$, $R_{1,6;4,8}$ and $R_{1,6;5,7}$ corresponding to the three successive pairs of complementary Au/Al contacts along the superconducting wire in figure 3. The shape of the $R$ versus $I_{dc}$ curve for $I_{dc} \leq 12 \mu A$ is similar to the $R$ versus $T$ curve for $T \leq T_c$. Near zero bias $I_{dc} = 0$, the resistance for all three curves is essentially 0, but rises monotonically until $I_{dc} \sim 12 \mu A$, except for a sharp peak at around 7.63 $\mu A$. This contribution to the differential resistance decreases as one goes further away from the FS interface. The similarity in shape in this current regime to the temperature dependent resistance shown in figure 4 suggests that this contribution is due to charge imbalance. If we scale the $R_{1,6;4,8}$ and $R_{1,6;5,7}$ curves, we can match the $R_{1,6;3,9}$ curve in the range $0 \leq I_{dc} \leq 12 \mu A$, as shown in figure 5(b). A fit of the inverse scale factors as a function of distance from the FS interface to the functional form in equation (5) gives a value for $\Lambda Q^*$ of 0.64 $\mu$m, in good agreement our rough estimate obtained from the temperature dependence in figure 4. It should be noted that the peaks at $I_{dc} = 7.63 \mu A$ in all three curves also match with these same scaling factors, indicating that they might be also associated with charge imbalance, although their origin is not clear to us at present.
At $I_{dc} \sim 12 \mu A$, there is a sharp decrease in the differential resistance of all three curves. All three curves show a negative differential resistance at this dc current. Measurements of the resistance of the superconductor (using the configuration $R_{1,6,9,3}$, for example) indicate that this value of $I_{dc}$ corresponds to the critical current of the superconductor, beyond which there is a transition to the normal state. Interestingly, the negative differential resistance peaks do not scale in the same way as the charge imbalance contribution, as can be seen directly from figure 5(b). In NS devices, this scaling is much clearer, as we shall show below. In some samples, there is a reflection of this negative differential resistance in the temperature dependence of the zero bias resistance. A hint of this behaviour can be seen in figure 4(b). In other samples, this behaviour is much more prominent, approaching 5% of the normal state resistance in some cases. The reason for the excess negative differential resistance is not clear, although it may be due to CAR.

4.1.2. Structures with two ferromagnets. In structures with a single ferromagnet, it is possible to inject a spin-polarized current into a normal metal or superconductor. With a second ferromagnetic element, a number of other experiments are potentially possible. For spin injection into a normal metal, the second ferromagnet can be used to detect the spin polarization as a function of distance from the injection point. Such spin-valve effects have been demonstrated recently by a number of groups in lateral mesoscopic structures [50]–[52]. For ferromagnets in contact with a superconductor, one may have correlated currents in the two FS interfaces resulting from CAR and EC as discussed above. Observation of EC and CAR in FS structures was recently reported by Beckmann et al [53]. We discuss below our own experiments on FS structures with two ferromagnetic elements.

Figure 6 shows a scanning electron micrograph of a Ni/Al/Au device with two elliptical ferromagnetic Ni particles in good (transparent) electrical contact with a superconducting Al wire. The separation of the two particles at their closest point is less than 100 nm, smaller than the superconducting coherence length of the Al. The two elliptical particles have different aspect ratios so that their coercive fields are different. Numbers refer to the contacts used in four-terminal measurements. The size bar is 1 $\mu$m.
Figure 7. (a) AMR of the left ferromagnetic particle of figure 6 at $T = 1.5$ K, when the superconductor is in the normal state. The magnetic field is aligned in the plane of the substrate along the major axes of the ferromagnetic ellipses. The coercive field of the larger left ferromagnetic particle is $\sim 185$ Oe, while the coercive field of the smaller right ferromagnetic particle is $\sim 305$ Oe. (b) Absence of any spin valve signal for the same sample at 1.5 K. Current is injected from the left ferromagnetic particle into the lower Al wire and the voltage is measured between the second ferromagnetic particle and the upper Al wire. (c) Resistance measured simultaneously across the two FS interfaces at 1.54 K. When the magnetizations of the two particles are aligned parallel, the resistance is higher, while when they are aligned antiparallel, the resistance is lower.
ferromagnetic particle has a similar AMR signature with a larger coercive field of 305 Oe as it has a longer aspect ratio.

With the information on the coercive field of each particle, one can manipulate the relative orientations of magnetizations of the two particles to be either parallel or antiparallel by simply varying the external magnetic field. Figure 7(b) shows the nonlocal resistance measurement of the device of figure 6 measured with the superconductor in the normal state \((T = 1.5 \text{K})\) as a function of magnetic field applied along the major axes of the elliptical particles. The current is sent from the left ferromagnetic particle into the lower Al contact, and the nonlocal voltage is measured between the second ferromagnetic particle and the upper Al contact. This is the configuration in which one expects to see a spin-valve signal, i.e. a difference in the nonlocal resistance depending on whether the magnetizations of the particles are aligned parallel or antiparallel, with the parallel configuration expected to have a larger resistance \([50]–[52]\). In our samples, no such spin-valve signal is observed. Instead, if we measure the resistance across both FS interfaces, as shown in figure 7(c), one sees a signature reminiscent of a spin-valve device: for example, as the external magnetic field is increased from \(-1200 \text{Oe}\), the resistance switches from a higher resistance to a lower resistance at the coercive field of the left ferromagnetic particle and then switches back to the higher resistance state at the coercive field of the right ferromagnetic particle. One may think that this behaviour is just a reflection of the AMR of each individual particle. However, if this as the case, one would observe a decrease in resistance before the coercive field of each particle is reached followed by a sharp increase in resistance, as seen in figure 7(a). This is clearly not what is observed in figure 7(c). We posit that the absence of a spin-valve signature in figure 7(b) in the classical spin configuration may be due to the small separation between the two ferromagnetic particles, which does not allow a spin-valve signal to develop sufficiently.

We now consider the case when the Al is superconducting. Figure 8(a) shows the temperature dependence of the resistance of the device of figure 6 in the measurement configuration of figure 7(c). Above the superconducting transition, one observes that the resistance of the parallel configuration is higher than that in the antiparallel configuration \(R_{\text{anti}}\), reflecting the data of figure 7(c). At the transition, \(R_{\text{par}} \sim R_{\text{anti}}\), but grows larger than \(R_{\text{anti}}\) as the temperature is lowered further, the difference in resistance becoming larger than even the resistance difference in the normal state. This is similar to the observations of Beckmann et al in their Fe/Al devices \([53]\), in which they also saw a difference between the resistance in the parallel and antiparallel configuration that was greater in the superconducting state than in the normal state, although the measurement in their experiment was in a nonlocal configuration, while figure 8 shows the resistance of the two FS interfaces in series in a local measurement configuration. Beckmann et al ascribed their observations to a spin-valve effect above \(T_c\) and CAR/EC effects below \(T_c\). From our perspective, while the effects below \(T_c\) may be due to CAR and EC, one cannot preclude a spin-valve effect below \(T_c\) with a different spin-diffusion length, and hence a different magnitude.

While our nonlocal measurements on this device show no difference in resistance between parallel and antiparallel configuration at zero current bias, we do observe differences in the nonlocal resistance between the two magnetization configurations at finite current bias. This is demonstrated in figure 9(a), which shows the nonlocal resistance as a function of dc current for the parallel and antiparallel magnetization orientations. Each of the two curves represents a difference between two local resistance measurements, \(R_{6.8;10,3} = R_{6.8;7.3} - R_{6.8;7.10}\). At zero bias, no difference is observed, but as the current bias increases, the resistance of the parallel configuration becomes larger than that of the antiparallel configuration. At a finite bias of around

New Journal of Physics 9 (2007) 116 (http://www.njp.org/)
Figure 8. (a) Temperature dependence of the device of figure 6 in the configuration of figure 7(c), when the magnetizations of the two ferromagnets are parallel and antiparallel. Parallel configuration taken in an external field of 0 Oe; antiparallel configuration taken in a field of 241 Oe. (b) Difference between the resistance of parallel and antiparallel configurations shown in figure 8(a).

23 µA, near the critical transition, the resistance in the antiparallel configuration is much lower than that of the parallel configuration, the resistances in the two configurations becoming the same as the current bias is increased further. This can be seen in figure 9(b), which shows the difference between the two configurations as a function of current bias. It is important to note that this behaviour in the differential resistance reflects the behaviour seen in the temperature dependence shown in figure 8.

It is tempting to ascribe the differences in the parallel and antiparallel resistances seen in figures 8 and 9 to CAR and EC. In the temperature dependence, for example, the resistance of the parallel configuration is larger than the antiparallel configuration (except near the superconducting transition). To recall the discussion given in section 2, the sign of the nonlocal resistance is positive for EC and negative for CAR; EC is favoured for the parallel magnetization configuration while CAR is favoured for the antiparallel magnetization configuration. However, the magnitude of the difference in resistance between the parallel and antiparallel configurations in our experiments and in those of Beckmann et al is very small. The fact that one also sees
Figure 9. (a) Difference of two differential resistance measurements of the NS interface, $R_{6,8;7,10} - R_{6,8;7,3}$, as a function of dc current, $I_{6,8}$, of the device of figure 6 when the magnetizations of the two ferromagnets are parallel and antiparallel. Parallel configuration taken in an external field of 0 Oe; antiparallel configuration taken in a field of 241 Oe. (b) Difference between the differential resistance of parallel and antiparallel configurations shown in figure 9(a).

a resistance difference above $T_c$ leaves open the possibility that the effect below $T_c$ may also be a spin-valve effect. In the case of the differential resistance (figure 9), the effect seems to be more likely to be related to superconductivity since the largest differences between the antiparallel and parallel configurations occur near the superconducting transition. However, it must be remembered that associated with each ferromagnetic element in these devices is a local magnetic field, so that the total magnetic field on the superconductor in the parallel and antiparallel configurations may not be the same even when the externally applied magnetic field is the same. It is therefore very hard to deconvolute the different potential contributions to the resistance difference in FS structures, especially since the effect is relatively small. It is for this reason that we have also measured NS structures without ferromagnets, where the effects due to CAR and EC are much larger. We discuss the results from these samples in the next section.
Figure 10. (a) Temperature dependence of the four-terminal resistance of the Au/Al cross shown in the inset to the figure. The current is sent from one Au into one of the Al leads, and the voltage is measured between the second Au wire and the second Al lead. (b) Four-terminal differential resistance of the same sample at 20 mK.

4.2. Superconductor/normal-metal structures

As an example of the magnitude of the nonlocal effects that can be observed in NS structures, figure 10(a) shows the temperature dependence of the four-terminal resistance of an Au/Al cross. Above $T_c$, the resistance is $70 \text{ m}\Omega$ which is comparable to the Sharvin resistance of the contact given its area, indicating that the interface between the Au and Al is very transparent. As the temperature decreases, a sharp peak in the resistance is observed near $T_c$, corresponding to the charge imbalance contribution we have seen already in the FNS samples. As the temperature is decreased further, however, the resistance does not vanish as in the FNS samples, but approaches a large finite value that is much greater than the normal state resistance. This behaviour is also seen in the differential resistance $dV/dI$ as a function of current $I_{dc}$ through the NS interface, shown in figure 10(b). Overall, the shape of the curve is very similar to the differential resistance of the FNS structures, except that the resistance at zero bias is large and finite, corresponding to the finite resistance at low temperatures seen in figure 10(a). This large, low-temperature, zero-bias contribution to the resistance is due to EC and CAR. If one were to be strictly accurate, the data shown in figure 10 do not correspond to the nonlocal CAR and EC contributions, since we are measuring the small but finite resistance of the NS interface. In order to measure the true nonlocal EC and CAR contribution, we have fabricated and measured devices similar to that shown in figure 11 [32]. The device consists of an Al wire in contact with numerous Au wires along its length. This device geometry enables us to measure the nonlocal resistance as a function of the distance from the NS interface through which current is being sent. Figure 12(a) shows the temperature dependence of the nonlocal resistance involving the four Au leads closest to the NS interface through which current is being sent. Since these are truly nonlocal measurements, the resistance for all four traces vanishes above $T_c$, and shows a peak in the resistance at $T_c$. $T_c$, however, is only approximately $600 \text{ mK}$, much less than typical for Al. This is because the transparent contacts between the Au and Al films result in a strong inverse proximity effect that suppresses the gap and $T_c$ in the Al. For the same reason, the resistance peaks are also considerably broader than seen in the NS cross. Nevertheless, the temperature dependence is
similar in form to what is observed in the NS cross: as the temperature is lowered, a peak in resistance is observed at $T_c$ that decreases as the temperature is lowered further, saturating at a finite value as $T \to 0$. Both the peak and the $T \sim 0$ resistance decrease in amplitude as one moves away from the NS interface through which the current is being sent. However, as we shall see shortly, the peak resistance and the $T \sim 0$ resistance do not scale with distance from the NS interface in the same way.

The low-temperature nonlocal differential resistance of the same device is shown in figure 12(b) for all six Au contacts, with the dc current sent between the Au wire labelled 2 to the Al wire labelled 1 in figure 11. Like the temperature dependence, the differential resistance is very similar to that of the NS cross. At zero dc current, there is a finite differential resistance that decreases as one moves further away from the NS interface through which the current is being sent. As the dc current is increased further, a large peak in resistance develops, followed quickly at around 2.75 $\mu$A by a region of negative differential resistance before the resistance eventually vanishes. From direct measurements of the resistance of the Al wire, this current corresponds to the critical current $I_c$ of the Al wire. The resistance peaks and corresponding resistance dips also decrease in amplitude as one moves away from the interface. As with the temperature dependence, the scaling of the peaks and the zero bias resistance with distance from the NS interface is not the same. This can be seen by scaling all the curves so that the amplitude of the peaks match, as shown in figure 13. If this is done, the zero bias resistances do not match. Interestingly, the magnitude of the resistance dips also do not scale in the same way as the resistance peaks, indicating that there is an additional contribution to the resistance near $I_c$ that is not associated with charge imbalance. A similar effect was seen earlier in our FS samples. The sign of the resistance dip indicates that this might be due to CAR. However, in the absence of a theory for nonlocal charge imbalance, it is difficult to distinguish between these contributions to the differential resistance.

Russo et al [54] recently measured nonlocal resistances in NS devices with tunnelling contacts. They also observed a positive differential resistance at zero bias that switched to a
negative differential resistance at finite bias before vanishing entirely as the bias was increased further. They ascribed the zero bias positive differential resistance to EC and the negative differential resistance to CAR, with the crossover bias at which the change from positive to negative current occurred related to the Thouless energy \( E_c = \hbar D / d^2 \), where \( d \) was the thickness of the superconductor in the experiments. As we noted above, earlier theoretical work indicated that in the tunnelling regime, EC and CAR should cancel exactly at all energies, leading to a vanishing nonlocal resistance. However, more recent theoretical work by Morten et al [37] predicts that EC should in fact be dominant for all interface transparencies, and that there also should be a dependence on \( E_c \), although the crossover from positive differential resistance to negative differential resistance at an energy \( E_c \) could not be described.

The data from our sample in figure 12(b) show that the crossover point from positive differential resistance to negative differential resistance does not depend on the distance between the NS interface and the second Au contact, since it is the same for all six Au contacts, i.e. the crossover point does not depend on \( E_c \). (While the data of figure 12(b) is shown as a function of dc current, the same conclusion is valid as a function of voltage bias, since the current goes

\[ \text{Figure 12.} \ \text{(a) Temperature dependence of the zero-bias nonlocal four-terminal resistance involving the four Au contacts nearest to the NS interface through which the current is being sent. The lead numbers refer to figure 11. (b) dc current bias dependence of the nonlocal four-terminal differential resistance involving all six Au contacts at } T = 20 \text{ mK. The dc current is sent from the Au lead labelled 2 to the Al lead labelled 1.} \]
Figure 13. Differential resistance curves of figure 12(b) scaled so that the resistance peaks at $\sim 2.75 \mu A$ match the height of the resistance peak of the Au lead closest to the NS interface ($R_{2,1;3,7}$). It must be noted that our devices show a large charge imbalance contribution that may make it difficult to discern a crossover from EC to CAR. However, the distance between the sixth Au contact and the NS interface is a factor of 6 larger than the distance between the first Au contact and the interface, corresponding to a factor of 36 in $E_c$. If there were a dependence of the crossover point on $E_c$, this should be clearly visible in the data of figure 12(b).

In fact, one can see data very similar to that seen by Russo et al if one looks at the differential resistance as a function of $I_{dc}$ at higher temperatures. Figure 14 shows the differential resistance of the first Au contact at a number of different temperatures from 20 mK to above the superconducting transition $T_c$. The trace at 500 mK in particular is remarkably similar to the data of Russo et al, showing a crossover from positive differential resistance at zero bias to negative differential resistance at higher bias without an intermediate peak in resistance. However, examination of the overall evolution of the traces with temperatures shows that this peak at zero bias is associated with charge imbalance: the charge imbalance peak that appears at higher bias at lower temperatures moves to lower bias as the superconducting gap diminishes with increasing temperature. It should be noted, though, that while the negative differential resistance of our 500 mK data is associated with the superconducting transition, this is not the case for the data of Russo et al where the energy scale associated with the differential resistance dips was much lower than the gap.

If charge imbalance can also contribute to the positive differential resistance at zero bias, how can we distinguish between EC, CAR and charge imbalance? As we have noted above,
Figure 14. Differential nonlocal resistance involving the first Au contact, $R_{2,1;3,7}$, at a number of different temperatures. The dc current is sent from the Au wire 2 to the Al wire 1 (see figure 11).

At low enough temperatures, the charge imbalance contribution should vanish at zero bias, leaving only contributions due to EC and CAR. Consequently, the zero bias conductance at low temperatures should show a length dependence that is distinct from the length dependence of the charge imbalance peak near $T_c$ in the temperature dependent resistance and the charge imbalance peak near $I_c$ in the differential resistance. Figure 15 shows the length dependence of the low temperature zero bias resistance and the charge imbalance peaks in the temperature dependence and differential resistance as a function of length from the NS interface, the data being taken from figure 12. Firstly, it should be noted that the scaling of the low temperature resistance from figure 12(a) maps the length dependence of the zero bias resistance taken from figure 12(b), and the scaling of the charge imbalance peak matches the scaling of the peak in the differential resistance. Secondly, without any analysis, it can be seen that the zero bias resistance decays on a length scale much shorter than the charge imbalance peak. If one fits the zero bias differential resistance data to an exponential decay of the form $A e^{-L/\xi_S}$, one obtains a value of $\xi_S = 315 \text{ nm}$. The superconducting coherence length for the diffusive Al in our group is typically of the order of 130 nm. However, the transition temperature of the Al wire in this sample is about a factor of 2 smaller than is typical, hence the value of $\xi_S$ we obtain is reasonable.

The charge imbalance peak is expected to decrease with the distance $L$ from the NS interface on a length scale determined by $\Lambda_{Q^*}$, as given by equation (5). A fit to this equation gives a value of $\Lambda_{Q^*} = 1038 \text{ nm}$. This is much smaller than the charge imbalance lengths found in early studies of charge imbalance in Al [43]–[45]; however, as we have remarked above, analysis of the magnitude of the peak in resistance due to charge imbalance in the temperature dependence in terms of equation (6) gives a value of $\Lambda_{Q^*}$ much more in line with the value from this fit. In
Figure 15. Length dependence of the zero bias differential resistance (filled circles), differential resistance peak (filled squares) and differential resistance dip (filled triangles) from figure 12(b), and the $T \rightarrow 0$ resistance (open circles) and peak resistance (open squares) from figure 12(a). All data are normalized to their values at $L = 210$ nm. The solid black line is a fit of the filled circles to the functional form expected for EC and CAR, $A e^{-L/\xi}$, with $\xi = 315$ nm. The dashed red line is a fit of the filled squares to the functional form expected for charge imbalance, $A[1 - \tanh(L/\Lambda)]$, equation 5, with $\Lambda = 1039$ nm.

In fact, a much better fit to the charge imbalance data in figure 15 is obtained if we use the linear length dependence represented by equation (6) instead of equation (5), with a fitting parameter of $\Lambda = 1445$ nm [32]. The use of equation (6) would imply that the Au contact measures the normal metal potential rather than the quasiparticle potential in the superconductor, but we do not know why this would be the case. Finally, we note that the length dependence of the resistance dips of figure 12(b) also shown in figure 15 exhibit a length dependence that decays slightly faster than the charge imbalance signal, as might be the case if their length dependence came from a combination of charge imbalance and EC/CAR.

5. Summary

CAR and EC are two processes that are predicted to occur in FS and NS structures that involve coherent coupling of quasiparticles in spatially separated normal metals or ferromagnets through their mutual interaction with a superconductor. The coherent coupling of the two quasiparticles in a pair can be demonstrated by tickling one of the quasiparticles in one normal metal or ferromagnetic electrode and observing the influence on the properties of the second normal metal or ferromagnetic electrode. The nonlocal transport measurements performed by all groups to date are in this category, where one drives a current through one of the NS or FS interfaces,
and measures the resulting nonlocal voltage on the second normal metal or ferromagnet. In the experiments performed in our group and the work of Beckmann et al and Russo et al, there is strong evidence for the existence of these nonlocal effects.

A number of issues remain to be resolved. The most critical is the ability to distinguish experimentally between EC and CAR. Using sample structures that incorporate two ferromagnets seems to be a very attractive means of distinguishing between EC and CAR, as one needs only to reverse the relative magnetization to switch between EC and CAR. However, as we have seen in our own experiments and those of Beckmann et al, the experimental signal in FSN structures is unexpectedly small, and complications from spin-valve effects and the magnetic fields generated from the ferromagnetic elements cannot be ruled out. In NS structures, the nonlocal effects are larger and complications from spin-valve effects and local magnetic fields are absent. In FS and NS devices with moderate to good interface transparencies, nonlocal charge imbalance is a prominent effect, and one for which no theoretical description is available. Other means of distinguishing between EC and CAR have been proposed such as looking at the correlations in the current noise between the two NS or FS interfaces [55], but no experimental results have yet been published.

In our opinion, it is important to have a theory of EC and CAR that can be used to describe the experimental results. In this regard, the most important aspect is to incorporate the effects of nonlocal charge imbalance within the same theoretical framework. To our knowledge, this has not been done. This would allow us, for example, to fit the experimental current and temperature dependence of the nonlocal differential resistance which is not possible at present.

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New Journal of Physics 9 (2007) 116 (http://www.njp.org/)
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