Resistance of normal-metal-dirty-superconductor interfaces

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Abstract. Measurements have been made of the resistive properties of a series of SNS sandwiches of In/W/In containing 0–10% Pb in In. The low-temperature interface resistance attributable to the alloying of the superconductor varied linearly with the resistivity of the InPb alloy, as predicted by Harding, Pippard and Tomlinson. The magnitude of this resistance was, however, found to be a factor of five less than predicted. The theory has been modified at higher temperatures, and agreement between theory and experiment for the form of $R_{SNS}(T)$ in dirty sandwiches has been obtained for the first time.

1. Introduction

There has been much work focused on the question of the resistance of normal–super-conducting (NS) interfaces in which s is a clean superconductor (e.g., Pippard *et al* [1], Battersby and Waldram [2]); the behaviour is fairly well understood, especially at temperatures very close to T_c. In contrast there has been only one systematic investigation of the properties of NS interfaces in which s was dirty (Harding *et al* [3]). Agreement between experiment and theory was found to be poor in this case.

Figure 1 shows typical resistance data obtained on an SNS sandwich in which the superconductors were clean. It can be seen that the resistance varies only slowly with T in the range up to 0.8 T_c. At these temperatures most of the excitations carrying the current in N will have energies much less than Δ, the bulk energy gap in s. At the interface most of these excitations are Andreev reflected [4] with electron–hole (e–h) inversion, a process which leads to no interface resistance. However, a small resistance is observed even at the lowest temperatures due to reflection without e–h inversion caused by mismatch between the two metals and dirt at the interface (these effects actually lead to a slow decrease in the resistance in the range up to 0.8 T_c as discussed in Lean and Waldram [6]. As the temperature is raised above about 0.8 T_c, significant numbers of excitations have energies greater than Δ. It was shown by Waldram [5] that these excitations generate a distribution of charge imbalance in s which decays exponentially as a function of distance away from the interface with a characteristic decay length denoted by λ_3. This charge imbalance corresponds to an electric field in s which is observed as an extra interface resistance. As the temperature is increased towards T_c, the charge imbalance relaxation processes in s become slower. λ_3 therefore diverges and this leads to the observed divergence in the resistance at T_c.
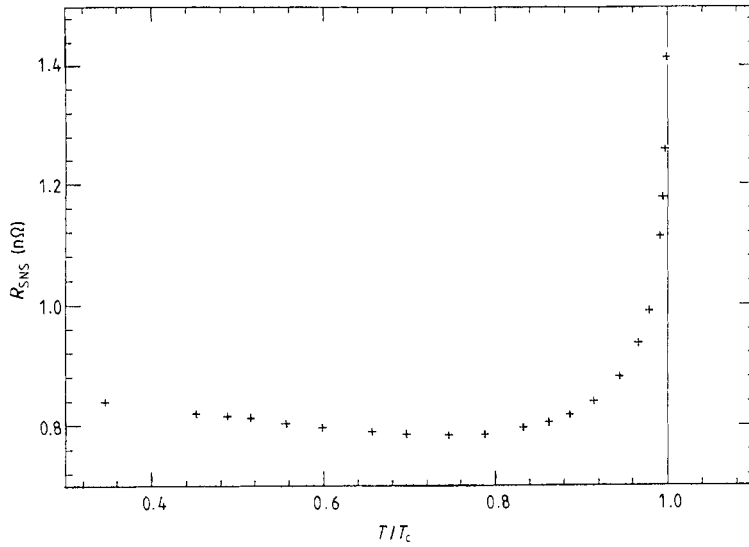
Figure 1. The $R_{SNS}(T)$ data obtained with a clean In/Ω/In sandwich.

Harding and co-workers found experimentally that alloying s altered the resistive properties of the interfaces in two ways, schematically illustrated in figure 2. Firstly the divergence in $R_{SNS}(T)$ observed at T_c was much larger in the dirty samples. Secondly the overall interface resistance at low temperatures is much larger in the dirty samples. Both these effects were easily explained at a semi-quantitative level. In both cases, however, discrepancies were found when attempts were made to fit the data to detailed theories. In this paper we describe a new investigation of the resistive properties of dirty SNS sandwiches which goes some way towards resolving these discrepancies.
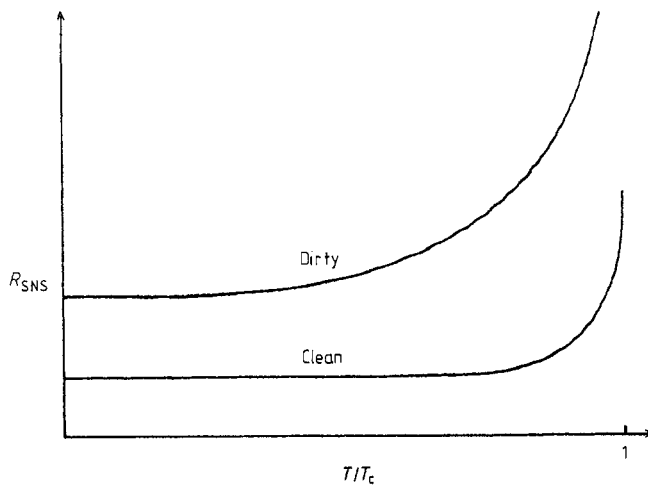

Figure 2. A schematic illustration of the effect of alloying s on the $R_{SNS}(T)$ curve.

2. Theory

Alloying s may change R_{SNS} through two mechanisms.

(i) Supergap excitations which enter s from N will be much more strongly scattered. As shown by Battersby and Waldram [2] this contribution to the interface resistance is expected to be proportional to $\rho^{1/2}$ where ρ is the residual resistivity of the superconductor. This is the reason why one expects the divergence at T_c to be much larger in the dirty samples.

(ii) Reflection of subgap excitations at the rise in Δ is no longer wholly Andreev reflection, i.e. some of the subgap electrons incident on the interface from N are expected to be reflected as electrons (rather than holes), causing resistance. This effect was first predicted by Harding *et al* [3] as a consequence of the scattering by impurities of the evanescent tails of the subgap excitations which extend into s. The result of the scattering is that parts of the evanescent tails return to the interface and cause some electron amplitude to be reflected back into N as well as pure hole amplitude. The phenomenon is analagous to frustrated total internal reflection in optics.

Our discussion of the theory of dirty NS interfaces will concentrate on (ii) since (i) is already included in the general resistance theory of Waldram [5] (and is clearly unimportant at low temperatures).

It is necessary to examine first the subgap boundary conditions for the interface which can be derived by solving the Bogoliubov equations for the excitations in the neighbourhood of the interface. Using the notation of Harding *et al* the Andreev reflection process can be represented by the formal statement.

$$e^+(1) + h^+(1) = A^+(1). \tag{1}$$

In this notation e and h are labels representing electrons and holes in N, A and B are labels representing the corresponding evanescent modes in s decaying and growing respectively in the direction of the wavevector k, and the superscript $+$ or $-$ represents the direction of k. The numbers in brackets give the phases of the waves in complex representation and the origin of coordinates has been chosen so that in (1) all the waves are in phase. (1) is a symbolic statement that an electron incident from N and the resulting reflected hole must be accompanied by an evanescent mode decaying into s. As mentioned above, it is the scattering of this evanescent mode by impurities which is central to the effect under discussion.

By examining the solutions to the Bogoliubov equations in the simple one-dimensional model of step function $\Delta(x)$ and no interface mismatch (Lean and Waldram [6]), it can be seen that it is also possible to combine electron and hole amplitudes in N with such phases that they will be accompanied by only a B^+ wave in s:

$$e^+(e^{i\varphi}) + h^+(e^{-i\varphi}) = B^+(1) \tag{2}$$

where $\varphi = \cos^{-1}(E/\Delta)$. (Harding *et al* incorrectly gave the hole component phase as $-e^{i\varphi}$. This makes no difference to the low-temperature resistance since if $E = 0$, $\varphi = \pi/2$. The phase factor is, however, of importance in discussing the form of $R_{SNS}(T)$, as we shall see shortly.)

We now retrace the argument given by Harding *et al* including the correct phase factor given by (2). We obtain the subgap boundary condition in the form

$$P(E) = (l_a/l_0)^S \sin^2(\varphi) \tag{3}$$

where $l_a = \hbar v_F / 2(\Delta^2 - E^2)^{1/2}$ is the amplitude decay length for the evanscent modes. $P(E)$ is the ratio of the normal and Andreev reflection probabilities, as introduced by Harding *et al*.

In order to calculate the resistance of the interface, we insert this boundary condition into the general theory derived by Waldram [5], using as supergap boundary condition the simple one suggested by Battersby and Waldram [2] that there is no probability of reflection of any type for the supergap excitations. This leads to the result:

$$Q_i = \frac{2f(\Delta) + I_p}{(l_0/\lambda_3)^S + (l_0/\lambda_2)^N (1 - 2f(\Delta) - I_p)} \tag{4}$$

where

$$I_p = 2[(l_0/\lambda_2)^N + (l_0/\lambda_3)^S] \int_0^\Delta -f'(E)P \, dE/(1 + P(l_0/\lambda_2)^N).$$

λ_2 is a diffusion length for all forms of inelastic scattering as defined by Waldram and λ_3 is the diffusion length for charge imbalance relaxation processes. In accordance with the notation of Harding *et al* the resistance is here given as Q_i which is the equivalent number of mean free paths in N. P may be rewritten as

$$P(E) = 2(E_S/\Delta)(1 - (E/\Delta)^2)^{1/2} \tag{5}$$

where $E_S = \hbar v_F / 2l_0$ and E_S is a parameter which gives the magnitude of the resistance generated by the scattering of the evanescent tails which is proportional to the resistivity of s. At low temperatures (4) and (5) reduce to the result quoted by Harding *et al*.

$$Q_p = (2l_a/l_0)^S. \tag{6}$$

Since l_a is approximately independent of impurity concentration, this implies that the interface resistance at low temperatures should be proportional to the residual resistivity of s.

The theory as described so far has been entirely one dimensional. Pippard [7] gave a three-dimensional calculation in which

$$Q_p = (l_a/2l_0)^S \qquad \text{(3D)}. \tag{7}$$

The extension to three dimensions reduces the magnitude of the resistance expected at low temperatures by a factor of four, but leaves the form of the relationship otherwise unchanged.

3. Experimental method

The system used for this work was an In/W/In sandwich having up to 10 at.% Pb added to the In. This is in contrast to the work of Harding *et al* who used Pb/Cu/Pb sandwiches with Bi added to the Pb. The experimental work aimed to measure the $R_{SNS}(T)$ curves of dirty samples and also the low-temperature interface resistance attributable to the impurity scattering of evanescent tails, as discussed in the § 2.

The second measurement was not easy since the resistance of interest was always much smaller than the overall low-temperature sample resistance (attributable to the resistance of the W slice and mismatch effects at the interface). The experimental procedure used was as follows.

(i) A thin (~0.25 mm thick) slice of zone-refined W was clamped horizontally in an evaporator and pieces of In placed on top. After pump-down the W was resistively heated to orange heat by passing a current of 100 A along the slice which cleaned the slice of surface contamination. It also caused the In to melt and spread over the entire upper surface of the slice. The interface thus formed was relatively free from contamination because the In coated the W while the latter was orange hot. After cooling, the evaporator was opened, the slice inverted and the procedure repeated to coat the other side of the slice.

(ii) The result of this process was a slice of W coated on both sides with In. This was next converted into a sample with interfaces of well defined area by clamping it between two short Pyrex tubes and heating the assembly in a small furnace. When the In melted, springs in the clamp forced the Pyrex tubes through the In until they were in contact with the W slice. After allowing the slice to cool, the In was removed from all of the W surface except for the area inside the Pyrex rings, first with a scalpel to remove the bulk and then by etching in HCl. This resulted in a W slice having a disc of In on each side with interface area equal to the internal cross sectional area of the Pyrex rings.

(iii) These discs were next extended into cylindrical casts of pure In ≈ 0.8 cm long, by once again clamping the slice between the Pyrex tubes, heating the clamp and adding In which melted inside the Pyrex tubes and so formed the casts. This was done in two stages. The In was always added to the uppermost tube and while making the second cast the first was supported from below by a stainless-steel screw which fitted into the tube.

(iv) The resistance of the pure sample as a function of temperature was measured with a SQUID voltmeter. The cryostat and method of measurement were the same as that described by Battersby and Waldram [8].

(v) After the cryostat had warmed up, the pure sample was removed and the casts cut down to half their original length. It was then returned to the Pyrex tube clamp used to make it and heated in the furnace to remelt the casts. When the casts had melted In/ Pb alloy was added to the uppermost one and the mixture stirred thoroughly. (The inverted cast was again supported with a stainless-steel screw.) This process was repeated to add the alloy to the other cast.

(vi) The resulting dirty sample was remeasured in the cryostat. The increase in the low-temperature resistance was predominantly caused by the low-temperature interface resistance of interest. Steps (i)–(vi) were repeated many times adding different quantities of alloy to obtain $R_{SNS}(T)$ and low-temperature resistance data as a function of alloy concentration.

An important check of this method was clearly to remelt the casts and add pure In to see if the recasting process in itself altered the resistance significantly. It was found that this typically increased the sample resistance by 0.015 nΩ which was satisfactorily small compared to the interface resistance, which for the alloy samples lay in the range 0.08–0.69 nΩ, as can be seen in figure 3.

4. Results and discussion

4.1. Low-temperature interface resistance

The data for the interface resistance at the lowest temperatures as a function of residual resistivity of s is shown in figure 3 and table 1. It can be seen immediately that there are
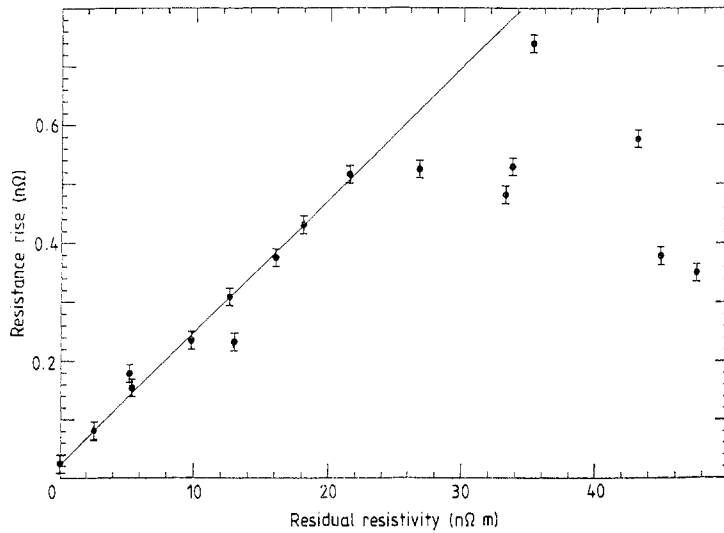
Figure 3. A rise in the low-temperature sandwich resistance on recasting as a function of residual resistivity of the PbIn alloy.

two regimes. Below $\rho \approx 25$ nΩ, which corresponds to ≈ 5 at.% Pb in the In, a linear relation can be seen, as expected from (6) and (7). Above this concentration the data becomes very irreproducible but the general trend appears to be that the interface resistance stops rising with concentration. The reason for this breakdown in the linear relation is not known but the irreproducibility strongly suggests that it is caused by some defect in the samples. For example, Harding *et al* found that his results became

Table 1. Samples used, showing the rise in R_{SNS} at the lowest temperatures caused by alloying, and other parameters.

Pb (at.%)	Sample no	ρ_{s} (nΩ m)	Resistance rise (nΩ)	T_{c} (K)
0	91	<0.01	0.024	3.41
0.5	115A	2.64	0.081	3.40
1	89A	5.46	0.154	3.42
1	79A	5.21	0.179	3.42
2	114A	9.73	0.234	3.48
3	97A	12.6	0.308	3.53
3	94A	12.9	0.232	3.53
4	76A	16.0	0.397	3.57
4.3	86A	18.0	0.431	3.60
5	85A	21.5	0.516	3.67
6	117A	26.8	0.524	3.79
7	113A	33.2	0.482	3.86
7	111A	33.7	0.530	3.87
7	99A	35.2	0.687	4.07
9	104A	43.1	0.577	4.32
9	98A	44.9	0.380	4.32
10	108A	47.5	0.350	4.39

irreproducible for concentrations for which his PbBi alloy was a two-phase system. This should not directly apply to the present data since the In/Pb system is thought to be a single phase up to about 13 at.% Pb [9]. It is, perhaps, possible that the interface favours and nucleates formation of the more concentrated phase above about 5 at.% Pb. Whatever the reason it is clear that the theory is not obeyed in the concentration region above 5 at.% Pb and in the rest of this paper we shall limit the discussion to data in the lower concentration range.

Below about 5 at.% Pb an encouragingly good linear relation is seen. However, when detailed calculations are made it is found that the magnitude of the measured slope is about a factor of five smaller than expected from (7). This result is to be compared with the conclusion of Harding *et al* whose data implied an interface resistance in Cu/PbBi about three times smaller than would be given by (7). The substantial difference between the two discrepancies suggests that they are related to some property of the pairs of metals involved rather than a systematic defect in the theory.

A likely cause of the discrepancy is mismatch between the Fermi surfaces of N and S. As discussed in [7] any difference between the size of the Fermi surfaces of N and S will cause the k-vectors of the excitations to change direction on crossing the interface. In addition excitations with some directions of k in S will be unable to enter N (a phenomenon analogous to total internal reflection in optics). The result of this is expected to be that less interface resistance will be seen since the excitations leaving S will have penetrated more deeply on average and will therefore have been more attenuated. In the case of Harding's Pb/Cu where the ratio of the Fermi surface areas s_{Cu}/s_{Pb} is 0.75, Pippard calculated that this effect should approximately halve the theoretically expected resistance. This was, however, still insufficient to bring theory into coincidence with experiment in that case implying that there was a further and unknown mechanism reducing the interface resistance. In the present case s_W/s_{In} is 0.68 so there is definitely appreciable mismatch. However, even a simple estimate of the size of the effect, as carried out by Pippard, is not possible since the Fermi surface in W is extremely complicated. It would seem likely, however, that Fermi surface mismatch is partly responsible for the observed discrepancy. Clearly more work needs to be carried out, ideally with different pairs of metals, to understand this discrepancy. (There are, however, problems with finding other possible pairs which do not alloy significantly with each other.)

4.2. Temperature dependence of interface resistance

Figure 4 shows $R_{SNS}(T)$ data for several samples with varying concentrations of Pb in the In. It can be seen, as would be expected from the heavier scattering of the supergap excitations, that the predominant feature is the increased size of the divergence at T_c. These curves have been fitted numerically to (4). l_0 was estimated from the resistivity of the casts above T_c using a value of ρl for W of 8.8×10^{-16} Ω m^2, which was based on the formula of Chambers [11] with the Fermi surface area of Sparlin and Marcus [12]. λ_2^N was estimated from the thermal conductivity data for W of Wagner *et al* [10]. The form of the temperature dependence of $\lambda_3(T)$ used was that of Battersby and Waldram [8]. The absolute value of this parameter was calculated from l_0 and the value of λ_3 required to fit the same sample area to the Battersby and Waldram theory, as described in Lean and Waldram [6] and Lean [13], using the fact that λ_3 is expected to be proportional to $l_0^{1/2}$. The two adjustable parameters were T_c, which was varied by a few mK to improve
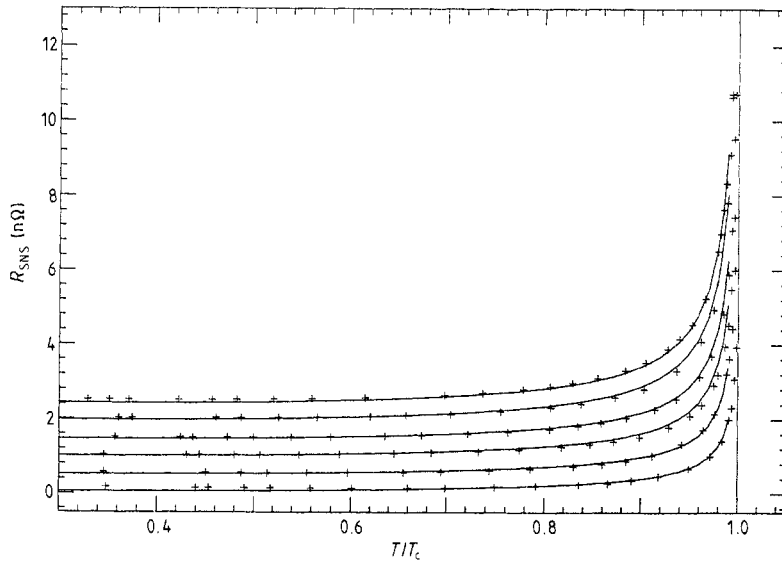
Figure 4. Fits of experimental $R_{SNS}(T)$ data to theory for dirty samples. The curves correspond to the entries in table 2 with the lowest curve representing the first entry (~0.5% Pb). Note that, for clarity, successive curves have been displaced vertically by 0.4 nΩ.

the fit to the divergence and R_W, the constant resistance which needed to be subtracted from R_{SNS} to give coincidence with the theoretical curve.

The value of E_S, which gives the magnitude of the effect due to the scattering of the evanescent tails, was obtained from the measured low-temperature behaviour. It should be noted that we have used the same one-dimensional expression (3) for the subgap reflection ratio P both in obtaining E_S at low temperatures and in fitting (4) at higher temperatures. This is not unreasonable since in (4) l_a and l_0 appear only as the ratio l_a/l_0. ($\sin^2\varphi$ is an energy-dependent factor.) The observed low-temperature behaviour implies that the dependence of P on l_a/l_0 has the form of (3) when l_0 only is varied if l_a is appropriately scaled. At high temperatures we are assuming that the scaling of l_a required to correct P for three-dimensionality, mismatch and any other causes of low-T discrepancy remains the same when T (i.e. l_a) is varied rather than l_0. The parameters are given in table 2.

The fits of the theoretical curves to the data are also shown in figure 4. The agreement between experiment and theory is generally good, though in the cleaner samples there

Table 2. Parameters of Pb/In alloys as a function of Pb concentration.

Pb (at.%)	(E_S/k_B) (K)	l_0 (10^{-8} m)	$(l_0/\lambda_3)^S$
0.5	0.91	21.6	0.116
1	1.85	10.6	0.078
2	3.37	5.83	0.060
3	4.34	4.50	0.052
4.3	6.42	3.16	0.045
5	7.40	2.65	0.040

are discrepancies between experiment and theory at low temperatures. These discrepancies correspond to the fall in R_{SNS} observed in clean SNS sandwiches between $0.3\ T_c$ and $0.8\ T_c$ due to imperfections of the interface. These effects have been neglected in the present theory and are the subject of Lean and Waldram [6].

It should be realised that the most important difference between our work in this section and that of Harding *et al* on $R_{SNS}(T)$ near T_c is the phase factor in (6). If the present theory is used without this factor a discrepancy between experiment and theory is seen similar to that found by Harding *et al*.

5. Conclusions

This work has confirmed in a new system the linear form of the low-temperature resistance of dirty SNS sandwiches as a function of impurity in s first observed by Harding *et al*, but only at low alloy concentrations. The magnitude of this resistance is a factor of five lower than that predicted by the theory of Pippard [7]. This discrepancy is not understood in detail but is thought to be partly caused by a Fermi surface mismatch. Once this discrepancy has been taken into account it is found that the form of $R_{SNS}(T)$ in the dirty sandwiches can be adequately explained by theory provided that the correct phase factors are used.

Acknowledgments

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