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Supercurrents in $YBa_2Cu_3O_{7-\delta}$–silver–indium junctions

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Abstract. We report detailed measurements of the properties of YBCO–silver–indium junctions produced on sintered YBCO. In almost all cases supercurrents are observed to pass through the junction below T_c of the indium. $I_c(T)$ is observed to be linear close to T_c and exponential at lower temperatures, consistent with proximity effect coupling exhibiting the usual decay length in the silver layer. However, the magnetic field dependence of I_c is consistent with many very small contacts rather than a uniform junction and the value of the contact resistance also implies that the contact obtained is imperfect.

1. Introduction

There has been much previous work on the question of making contacts to high-T_c superconductors. Such work is clearly important from the technological point of view but it is also to be hoped that the detailed study of the properties of YBCO/normal metal interfaces may increase our understanding of the physics of these materials. For example, one important issue is the proximity effect whose existence in normal metals in contact with YBCO has still not been conclusively demonstrated.

Most of the previous work on making contacts to sintered material (e.g. Van der Maas *et al* 1987) has involved depositing silver or gold onto the surface by some method (sputtering, evaporation, silver paint etc) and reannealing in oxygen. Contact resistivities in the range $10^{-7}\ \Omega\ cm^2$ have usually been obtained. These compare with values of $10^{-10}\ \Omega\ cm^2$ which can be obtained for normal-superconductor interfaces with conventional superconductors (see, for example, Lean and Waldram 1989) although it is not clear whether the 'ideal' contact resistivity to YBCO would be expected to be as low as this. However, work with YBCO single crystals (Jing *et al* 1989) where contact resistivities of order $10^{-9}\ \Omega\ cm^2$ have been observed implies that the presently produced contacts to sintered materials do suffer from unexpectedly high resistance. There have been claims (Ekin *et al* 1988) of $10^{-10}\ \Omega\ cm^2$ contact resistivities to sintered YBCO with essentially the preparation technique described above. These results may be in error, however, due to the incorrect assumption that the thin film silver contact was an equipotential (this issue has been termed the "spreading resistance" problem by Jing *et al* (1989).

In this paper we present some results on sintered YBCO/silver/indium junctions, where the YBCO/silver junctions were made by essentially the method used by previous workers. These observations were made in the course of establishing techniques to be

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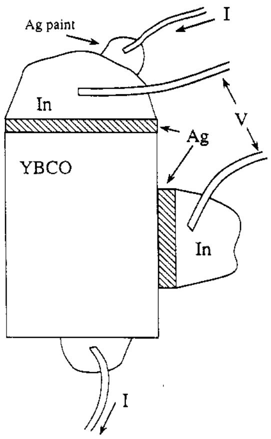

Figure 1. Sample arrangement.

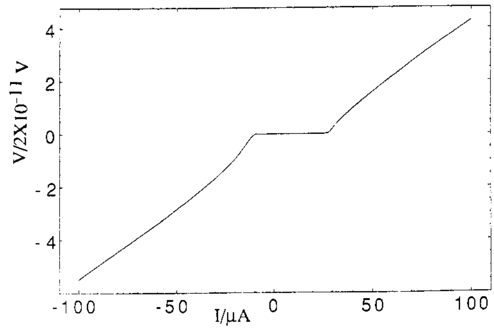

Figure 2. Typical *I–V* characteristic showing supercurrent

used for single crystal and epitaxial film samples on which we shall be reporting soon, and are to some extent preliminary. Nevertheless, we hope to show that our observations on the supercurrents which pass across the junction at temperatures less than T_c for the indium shed a useful light on the nature of the contact. Our observations also have implications for the existence of a proximity effect since, although the details of the junction may be unclear, it seems likely that the supercurrents are passing through at least a thin layer of silver.

2. Experimental details

The experiments used bars of cross section 2 mm^2 and about 5 mm long which had been cut from larger pellets of sintered YBCO. These bars were placed in an evaporator with a base pressure of 10^{-6} mbar, and a layer of silver about 1 μm thick was thermally evaporated onto the end. During this process the pressure usually rose to around 10^{-5} mbar. At an early stage in this work various attempts were made to improve the YBCO/Ag interface by cleaning the surface of the YBCO before evaporating the Ag. These methods included ion etching in both Ar and O_2 and produced only a marginal improvement in contact resistance. It was later found that the contact resistance was also only slightly improved by actually breaking the bar while evaporating so that a fresh surface was used, suggesting that no amount of cleaning was likely to improve matters. No surface cleaning was used in the samples used for the present measurements. After the evaporation the vacuum was broken and the sample annealed in flowing oxygen at 600 °C and then cooled to room temperature over 4 h. The indium/silver interface was formed by melting a cut piece of indium shot onto the sample, enclosing the voltage lead (figure 1). A voltage controlled soldering iron was used and care was taken not to heat the sample more than necessary. Since the measurements are made with a SQUID voltmeter the resistance of the voltage leads must be kept as low as possible. This was achieved by using superconducting leads and attaching the second voltage lead with a second YBCO/Ag/In structure fabricated at the same time as the first. The current leads were attached with silver paint.

It should be particularly noted that this arrangement, with a macroscopic piece of indium soldered onto the silver, avoids the 'spreading resistance' problem mentioned earlier: the silver can justly be assumed to be an equipotential. A price to be paid for this is that the silver/indium interface has to be measured in series with the silver/YBCO interface; however, the resistance of the silver/indium interface should be negligible compared to that of the YBCO/silver one. A test with an In/Ag/In junction did in fact show that this was the case: it was found to have a resistance two orders of magnitude below that of the YBCO/Ag/In junctions. A second problem with this approach is the possibility that the heating (to about 150 °C) while melting on the indium may change the YBCO/Ag interface in some undetermined way. In the configuration of this experiment it was not possible to avoid this difficulty by using pressed In contacts because of the high resistance which then resulted at the Ag/In interface. However, we have some indirect evidence that heating does not affect the YBCO/Ag interface drastically; in some later work on thin film samples using a different sample configuration we have found that samples with pressed In contacts have contact resistances of the same order of magnitude as those with melted In. It was in any case felt that this uncertainty was preferable to that of attempting to model the spreading resistance correction.

The I–V characteristics of the samples were measured at temperatures between 1.2 and 7 K using an RF SQUID in the conventional voltmeter configuration. The cryostat also contained a small solenoid to enable fields $\boldsymbol{B}$ of up to about 10^{-4} T to be applied to the sample in a direction along the plane of the silver film.

3. Results

3.1. Temperature dependence of I_c

The contact resistivity of the samples was found to be of order $10^{-5}\ \Omega\ \mathrm{cm}^2$ without annealing in oxygen and $10^{-7}\ \Omega\ \mathrm{cm}^2$ after annealing. These are about the values that one expects by comparison with previous work.

Out of about 20 samples measured, all but one have displayed supercurrents below the critical temperature T_c of the indium (figure 2). We have compared the temperature dependence of I_c with the predictions of Clarke (1969), based on de Gennes (1964) for the supercurrents in SNS junctions which gives:

$$I_c \propto \frac{\Delta_1}{(1 + \xi_{S1}/\xi_N)} \frac{\Delta_2}{(1 + \xi_{S2}/\xi_N)} \xi_N^{-1} \exp(-a/\xi_N). \qquad (1)$$

Here ξ_N is a decay length for the order parameter in the silver. It is essentially the range of the Gor'kov kernel, and in dirty material is expected to be of order $(hv_F l/kT)^{1/2}$. ξ_S is the Ginzburg–Landau coherence length in the superconductor. The suffixes 1 and 2 refer to the two superconductors, Δ is the equilibrium gap parameter deep inside the superconductor, and $\Delta/(1 + \xi_S/\xi_N)$ is an estimate of the gap at the superconductor-normal interface, which is reduced by the proximity effect. (This is a slight modification of the original expression of Clarke intended to allow for the possibility that $\xi_S < \xi_N$.) The exponential term arises through de Gennes' one-frequency approximation which implies that the order parameter decays exponentially into the normal metal from the SN interface and a is the thickness of the silver layer.

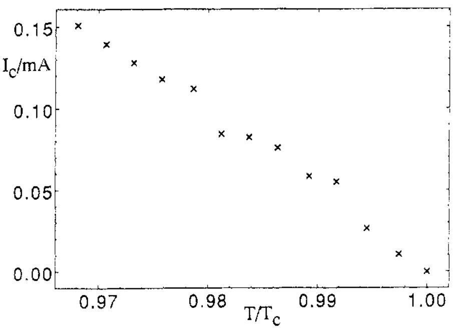

Figure 3. Typical plot of I_c against reduced temperature close to T_c. The range of temperatures shown corresponds to about 0.1 K below the indium transition temperature.

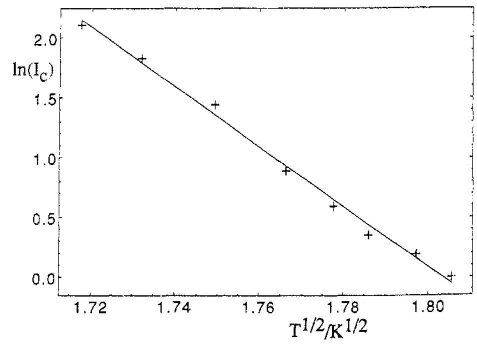

Figure 4. Typical plot of ln (I_c) against $T^{1/2}$ in the lower temperature range (in this case 2.95–3.25 K).

In the present case we have $\xi_{YBCO} \ll \xi_N$ and (1) simplifies to:

$$I_c \propto \Delta_{YBCO}\Delta_{In}(\xi_N + \xi_{In})^{-1}\exp(-a/\xi_N). \qquad (2)$$

It should be noted that this simplified treatment may give an incorrect account of the boundary condition on Δ is at the YBCO/Ag interface. Fortunately this does not affect the present analysis if, as seems likely so far below T_c of YBCO, a more correct treatment would introduce no additional temperature dependence. Expression (2) has been fitted to our measured values of $I_c(T)$. It is instructive to examine the behaviour close to T_c since previous workers (for example Clarke 1964) have investigated $I_c(T)$ in that range in both SNS and tunnel junctions. Sufficiently near T_c we have $\xi_{In} \gg \xi_N$, the factor $\Delta_{YBCO}\exp(-a/\xi_N)$ may be treated as constant and I_c should vary as Δ_{In}/ξ_{In}, which should be proportional to $1 - T/T_c$. This prediction is well obeyed for $T/T_c > 0.97$ (figure 3), providing some evidence that we are indeed seeing a proximity effect. If, for instance, the critical currents had been passing through indium–YBCO tunnel junctions formed in pinholes in the silver layer, $I_c(T)$ would have been expected to vary as Δ_{In}, which should be proportional to $(1 - T/T_c)^{1/2}$.

At lower temperatures the exponential term would be expected to dominate so we expect to see $I_c \propto \exp(-a/\xi_N) = \exp(-AT^{1/2})$ where A is a constant. We have estimated ξ_N from the dirty limit expression given above, the value of v_F used being that tabulated by Kittel (1963) and the mean free path being estimated from measurements of the resistivity of the silver films. The result of this estimation was $\xi_N T^{1/2} \approx 2.4 \times 10^{-7}$ mK$^{1/2}$. A typical plot of $\ln(I_c)$ against $T^{1/2}$ for lower temperature data is shown in figure 4 (the temperature range of these plots was somewhat limited because critical currents larger than about 9 mA were difficult to measure in the present apparatus because of excessive heating). In the range concerned the temperature dependence appears to be as predicted. The experimental value of the constant A together with the nominal value of the thickness of the silver, $a = 10^{-6}$ m, allows an estimate of $\xi_N T^{1/2}$ to be made. The measured values of $\xi_N T^{1/2}$ fall in the range $(0.8\text{–}2.0) \times 10^{-7}$ mK$^{1/2}$. Although this is rather smaller than the calculated value the agreement may be considered reasonable in view of the uncertainties in the value of a and in the calculation of ξ_N.

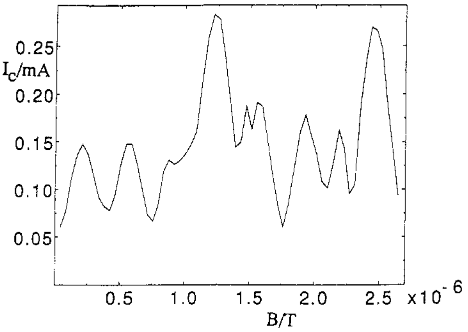

Figure 5. Small part of an $I_c(B)$ curve.

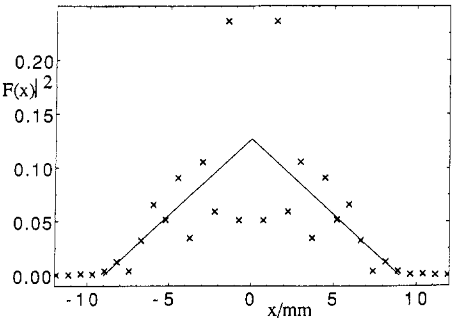

Figure 6. $|F(x)|^2$ calculated from data in figure 5.

3.2. Results for $I_c(B)$

An important test of the uniformity of the junction is the effect on I_c of the application of a field $\boldsymbol{B}$ parallel to the silver film. If a field is applied to a uniform junction whose width normal to the field is small compared to the Josephson penetration depth the resulting form of $I_c(B)$ should be a single slit Fraunhofer diffraction pattern. Calculations show that the junctions in the present work would be expected to become self field limited when I_c exceeded about 8 mA which was very rarely the case in the present data due to the heating problem mentioned in the last section. However, the application of a field to the present junctions resulted in a randomly oscillating $I_c(B)$ curve with the height of the peaks and troughs very variable and no definite period. No central peak was observed in the pattern and no falling envelope could be seen out to at least 500 periods. A small part of such a curve is shown in figure 5. Such apparently random curves are in general reproducible, but are changed irreproducibly by raising the temperature above the T_c for indium or by applying a field of more than about 10^{-4} T. This suggests strongly that we have flux trapped in the junction, which is randomising the phase of different regions. Our cryostat incorporates concentric mu-metal cans and a set of Helmholtz coils which between them should reduce the field at the sample to about 10^{-8} T, but this low field still corresponds to hundreds of flux lines penetrating the YBCO in the direction perpendicular to the film. If these flux lines are trapped in the YBCO when it is first cooled and subsequently compressed or partly compressed into the silver by the Meissner effect in the indium, our observations of $I_c(B)$ are understandable as follows.

We model the interface as a large number of small individual contacts of characteristic width t, and suppose that there is enough trapped flux present to make the phases of the individual contacts random in zero applied field. This explains why there is no central peak in the $I_c(B)$. When an external field is applied, the phases of the individual regions change, but they remain random, so $I_c(B)$ does not change in any systematic way with B. If this model is accepted one may make two deductions at once from the nature of the $I_c(B)$ curve. It is obvious that it contains structure on a scale down to about 2×10^{-7} T but no finer structure. A field of 2×10^{-7} T injects one flux quantum into a silver film of thickness 1 μm and width 1 cm. Bearing in mind uncertainties such as the compression of the extended field into the film by the superconducting indium and the thickness of the film, this observation strongly suggests that superconducting contacts exist over the full 2 mm width of the junction: if the junction had been narrower we should have needed a larger external field to make appreciable changes in the relative phases of the different regions. Secondly, since there is no reduction in the envelope up to fields 500 times larger than this, we may deduce that the width t of the individual contacts cannot be greater than 2 mm/500 = 4 μm. Since it seems unlikely that we have enough trapped flux-lines present to divide the interface into such small phase coherent regions, the observation suggests that the interface contains many, isolated contacts of width less than 4 μm.

One may check this model more quantitatively by Fourier analysis. We should have

$$I_c(B) = \left| \int J(x) \exp(ikx)\,dx \right| \tag{3}$$

where $J(x)$ is a complex supercurrent density per unit width which has been given a random phase by the trapped flux, and k is $2\pi Ba/\Phi_0$ where a is the magnetic thickness of the junction. Because $I_c(B)$ does not contain phase information, we cannot deduce $J(x)$, but by taking the transform of $I_c^2(B)$ we obtain $F(x)$ the autocorrelation function of $J(x)$. $F(x)$ is also complex, and it is best to consider $|F(x)|^2$. Taking a model of small contacts of random phase spread uniformly over a junction of width w it is easy to show that except for $x < t$ one expects $|F(x)|^2$ to be a random function with exponential distribution at each value of x, the mean of the distribution being a triangular function of x, with width $2w$. For $x < t$ the phases cease to be random, leading to a large central peak.

Figure 6 shows that our data appear to fit this model. The width of the junction obtained from the Fourier analysis is again of the correct order, although rather large at about 9 mm. The solid line in the figure represents an estimate of the triangular mean function calculated assuming the exponential distribution. The central peak is too high to be conveniently plotted. Its height, however, is related in a simple way to the range of B over which $I_c(B)$ was Fourier analysed and gives no new information about the sample.

4. The nature of the interface

As explained above, the observed $I_c(B)$ dependence is consistent with many small and isolated contacts in random positions and with random values of individual critical current. In what ways might this arise?

(i) The supercurrent might be flowing through pinholes in the silver, perhaps formed by shadowing on the microscopically uneven surface during evaporation. This seems unlikely for two reasons. Deliberately prepared indium-YBCO junctions have very high resistance, presumably because the indium reacts with the YBCO; and this model would not explain the encouraging fit to proximity effect theory.

(ii) The supercurrent might be flowing through a number of abnormally thin silver regions. This, however, would not explain the fit of the proximity theory for a silver thickness of 1 μm, nor would it by itself explain the high contact resistance.

(iii) There may be relatively few superconducting contacts between the grains of YBCO below the surface, even if the silver makes good contact to every surface grain. This model could explain the observations, but it seems implausible for good quality high-density materials (and would not explain similar results on epitaxial films which we will be reporting on later).

(iv) There may be few places where the silver–YBCO interface is clean. This is certainly a possible, perhaps the most likely explanation. Nevertheless, extensive efforts to clean the interface have all failed, and electron-microscope examination of epitaxial YBCO–silver interfaces deposited *in situ* on which we shall report later show clean interfaces, so there does not appear to be any intermediate compound which automatically precipitates out at the interface.

(v) There is evidence (McConnell and Morris 1989) that the silver diffuses down the YBCO grain boundaries during reannealing. Possibly the only good contacts are formed in this way.

(vi) It is possible that the YBCO electronic structure prevents good electrical contact unless the *ab* planes happen to be normal to the interface. Depending on the precise orientation requirement, this may only happen for relatively few grains.

We are inclined to reject the possibilities (i)–(iii). Which of the other models is correct may become clearer when we have more comparisons with the results on epitaxial films and single crystals of YBCO.

5. Conclusions

It is clear from this preliminary work that, in spite of strenuous efforts, we have only succeeded in making small, isolated contacts between the silver and sintered YBCO (though these small contacts do normally span the full width of the junction). The reasons for this are not yet clear. We are in the process of repeating this work with high-quality YBCO films of various orientations for which microscopic examination and characterisation of the interfaces is a good deal easier: this seems to us the best way to proceed in obtaining insight into both the fundamental physics and the technological contact problem.

Our work has also made obvious the confusing effects of even a seemingly small amount of trapped flux. It is interesting to note that one group (Akoh *et al* 1989) who have seen supercurrents showing the expected Frunhoeffer pattern in $I_c(B)$ used junctions considerably smaller than ours, but do not report how carefully they compensated stray fields.

In spite of these difficulties we believe that our quite successful fit of $I_c(B)$ to equation (1) both near T_c, and at lower temperatures where the measured value of $\xi_N T^{1/2}$ agreed reasonably well with the theoretical estimate for silver, provide encouraging evidence of a proximity coupling of YBCO to indium through silver which obeys the conventional theory, suggesting that the order parameters in the two superconductors have compatible symmetries.

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References

Akoh H, Shinoki F, Takahashi M and Takeda S 1989 *IEEE Trans. Magn.* **MAG-25** 79
Clarke J 1969 *Proc. R. Soc.* A **308** 447

Ekin J W, Larson T M, Bergren N F, Nelson A J, Swatzlander A B, Kazmevski L L, Panson A J and Blankenship B A 1988 *Appl. Phys. Lett.* **52** 1819
Jing T W, Wang Z Z and Ong N P 1989 *Appl. Phys. Lett.* **55** 1912
Kittel C 1963 *Introduction to Solid State Physics* 2nd edn (New York: Wiley) p 250
Lean H W and Waldram J R 1989 *J. Phys.: Condens. Matter* **1** 1285
McConnel M and Morris W 1990 *Proc. Boston Mater. Res. Soc. Conf. 169 (Boston, MA, 1989)*
Van der Maas J 1987 *Nature* **603** 328