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A Simple Method for Attaching Electrical Leads to Small Samples of High-T_c Oxides

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A simple technique for attaching a number of electrical leads to a small sample of $YBa_2Cu_3O_{7-x}$ has been devised. Thin gold wires are spark-bonded onto a crystal by using a capacitor discharge. As-bonded contacts are highly resistive suggesting a local oxygen deficiency caused by the bonding process. Contact resistance (for a contact area $\sim 10^{-2}$ mm^2) can be lowered to $\sim 1\ \Omega$ by annealing at 800°C in an oxygen atmosphere. Some results of electrical measurements on single crystal samples of $YBa_2Cu_3O_{7-x}$ using this technique are presented.

KEYWORDS: high-T_c oxides, electrical contact, spark-bonding, anisotropic resistivity

From the very early stage of the current boom of the high temperature superconductivity research, it has been recognized that making electrical contacts is by no means a trivial part of the experimental research when one attempts a transport measurement on this class of oxide materials. In fact, at least some of the "discoveries" of so-called super-high T_c's may be traced due to ill-defined electrical contacts. The most commonly employed method for achieving electrical contacts (because of its quickness and reasonable reliability) is the use of conducting paste or epoxy, however metallization by vacuum evaporation or sputtering generally gives more reliable contacts. A few other methods reported thus far include use of pressed indium contacts,[1] ultrasonic soldering,[2] high-temperature metallization of silver epoxy[3] and ultrasonic bonding.[4]

An electrical contact poses a particularly serious problem when one attempts to carry out transport measurements on single crystals, since only small single crystals are presently available. In this letter, we describe a simple technique for attaching a number of electrical leads on a small single-crystal sample. The method essentially consists of spark-bonding of gold wires and subsequent oxygen treatment. The spark-bonding itself is a time-honored technique used for metals and alloys and for semiconductors such as Ge. It has also been proven to work for the new high-T_c oxides.

The present method provides reliable electrical contacts with merits such as (1) mechanical strength, (2) a small and well-defined contact area, (3) low contact resistance, and (4) usability from low to very high temperatures. Some results for the anisotropic conductivity of $YBa_2Cu_3O_{7-x}$ single crystals obtained by this method are also presented.

Typical dimensions of the single crystals used for the transport measurements were $\sim 1\times 1\times 0.4$ mm^3, the largest face being perpendicular to the c-axis. The details of crystal growth and the superconducting characteristics of these single crystals are given in refs. 5 and 6. In order to measure the resistivity along the c-axis as well as that in the ab-plane, we generally attached eight leads, i.e. four on each of the two largest faces. Although just four leads suffice in principle, to carry out measurements of the anisotropic conductivity by, for example, the Montgomery method, this kind of redundancy enabled us to make a cross check of the data. We feel that cross-checking is absolutely important when one deals with highly anisotropic and possibly inhomogeneous samples. (In one case, we attached a total of fourteen leads on a $2.3\times 1.0\times 0.4$ mm^3 crystal to do measurements of the anisotropic conductivity as well as the Hall effect with full cross-checking.)

Figure 1 is an illustration of the set up used. Spark-bonding can be done using such a simple circuit composed of a battery and a capacitor. It is useful to be able to select from several capacitances to be selected according to the diameter of the gold wire and the surface condition of the sample. We typically used a 1 mF capacitor charged by a 9 V battery to spark-bond 50 μm wires. The following is a step-by-step process followed when we attached a number of leads on both faces of a small single-crystal sample.

(1) The sample is fixed on a piece of metal with silver paste only on one face of the sample. The metal is electrically connected to one terminal of the capacitor.

(2) A 50 μm-diameter gold wire is held with a pair of tweezers which is electrically connected to the other terminal of the charged capacitor, and is brought in contact with the sample under an optical microscope. Under

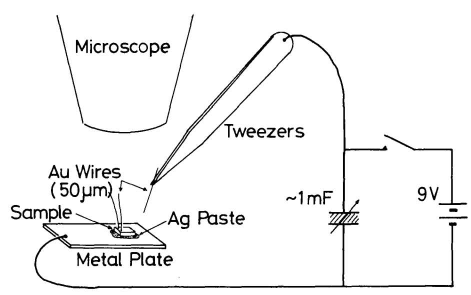


Fig. 1. A simple set up for spark-bonding of gold wires on a small sample of $YBa_2Cu_3O_{7-x}$. The details of the method are described in the text.

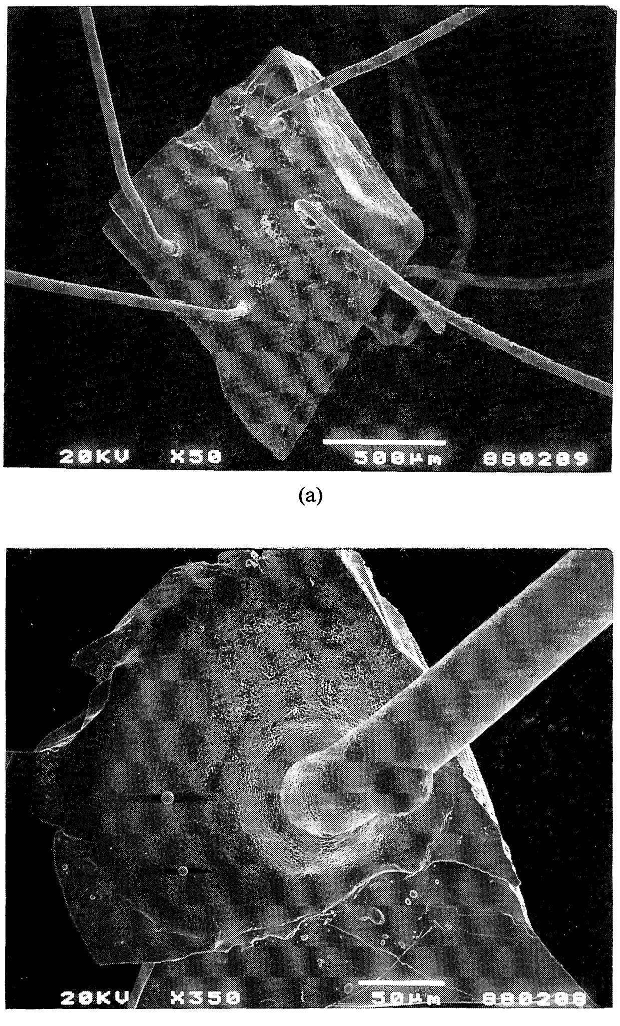
suitably adjusted conditions found by trial and error, a spark develops upon contact and the wire is bonded on the spot.

(3) After the required number of leads are attached on the face by repeating step (2), the silver paste is washed away.

(4) Steps (1) to (3) are repeated to attach leads on the other face of the sample.

With some training, one can routinely attach many leads on a small sample.

Figure 2(a) is a scanning electron micrograph of a sample with four leads on each of the ab-faces. The mechanical strength of the bonding is excellent. In fact, when we pull the wire too hard, a small part of the crystal around the contact is ripped off (or the wire breaks). Figure 2(b) shows such a part ripped off from a larger crystal. As is evident from this micrograph, local melting of the region near the contact has taken place during the spark-bonding process and the molten $YBa_2Cu_3O_{7-x}$ has wetted the gold wire. The region of the local melting appears to extend roughly 100 μm in diameter.



(a)

(b)

Fig. 2. (a) An $YBa_2Cu_3O_{7-x}$ single crystal with eight electrical leads attached (four are on the back). The white bar in the lower part of the photograph represents 500 μm. (b) Scanning electron micrograph of the region around a spark-bonded wire. The diameter of the gold wire is 50 μm. This is a part of the crystal which was ripped off when the wire was pulled hard.

As-bonded contacts are, however, highly resistive. The contact resistance is typically 10 to 100 kΩ at room temperature and shows a semiconductive temperature dependence. Obviously, the spark-bonding process has caused local damage (probably a severe oxygen deficiency), and the region around the contact has turned to a semiconducting phase. Although the attached wires are incomplete as electrodes at this stage, one may elect to make use only of their mechanical strength and use a small amount of conducting paste around the bonded area to achieve electrical contact.

Instead of resorting to such measures, one can simply lower the contact resistance by annealing in an oxygen atmosphere and make full use of the present method. Figure 3 shows the temperature dependence of the contact resistance measured *in situ* by a three terminal method during the oxygen annealing process. The annealing sequence for this sample is given in the figure. The inset shows an Arrhenius plot of the temperature dependence of the contact resistance. The as-bonded contact shows an activated temperature dependence with an activation energy of $\sim$0.35 eV. Annealing at 500°C did not change the semiconductive temperature dependence in a fundamental way, although the activation energy was lowered to $\sim$0.2 eV. Annealing for 8 h at 700°C yielded no significant improvement over that at 500°C. However, annealing at 800°C drastically changed the character of the contact. The temperature dependence turned metallic and the contact resistance at room temperature became only a few ohms. With an estimate of the contact area, 10^{-2} mm^2, this corresponds to $\sim 10^{-4}$ Ωcm^2.

It is known that at temperatures close to $\sim$1000°C,

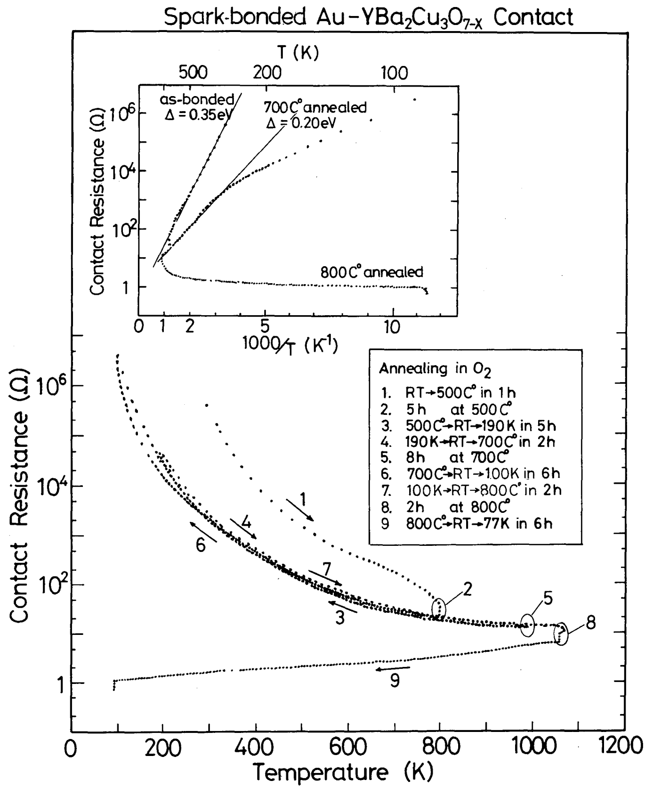


Fig. 3. Temperature dependence of the contact resistance. The numbers represent the annealing sequence. The inset is an Arrhenius plot showing activated behavior of the contact resistance.

decomposition of $YBa_2Cu_3O_{7-x}$ takes place[5,7] according to the reaction,

$$2YBa_2Cu_3O_{7-x} \longrightarrow Y_2BaCuO_5 + 3BaO + 5CuO + (0.5-x)O_2,$$

where Y_2BaCuO_5 (green phase) is an insulator. Although a SEM-EPMA study did not reveal any macroscopic segregation of the green phase, it is probable that this kind of decomposition reaction has occurred on a microscopic scale in the region around the contact, since it has experienced local melting. It is noted that the annealing temperature of 800°C is necessary to recover a metallic contact resistance. In a run similar to that shown in Fig. 3 for another sample, we found that the effect of annealing at 750°C was essentially the same as that at 700°C. The annealing temperature of 800°C seems to be higher than what would be required if the local "damage" were a simple oxygen deficiency. We speculate that a "damaged" region consists of a microscopically segregated mixture of various phases, and that the annealing process involved in the improvement of the contact resistance is essentially a partial recovery of the metallic phase and a formation of conducting paths by percolation.

Figure 4 shows an example of the resistivity data for one of our best quality crystals exhibiting a sharp superconducting transition with the zero resistance T_c of 91 K and the transition width less than 1 K. (It was taken from the same lot as those reported in ref. 6. For this sample, both the ab-plane resistivity ρ_{ab} and the c-axis resistivity ρ_c show linear temperature dependences. This is in contrast to recent reports[4,8,9] that the temperature dependence of ρ_c is semiconductor-like. The temperature dependence of ρ_c is currently an important issue because the linear temperature dependence of ρ_{ab} and σ_c is proposed by Anderson and Zou[10] to be an experimental evidence for the resonating valence bond state.

We observed a semiconductor-like temperature dependence of ρ_c for the samples, whose quality was not as good as the one shown in Fig. 4 with respect to the superconducting characteristics. We even experienced a case in which different combinations of electrical probes on a single sample gave different temperature dependences of ρ_c. It is thus difficult at the moment to judge whether the metallic or the semiconductor-like temperature dependence represents the intrinsic behavior of ρ_c. Thus the present study clearly demonstrates the need for more careful measurements with full cross-checking; the method described in this letter provides a powerful tool for such studies. Detailed discussion of the transport measurements will be presented in a separate paper.

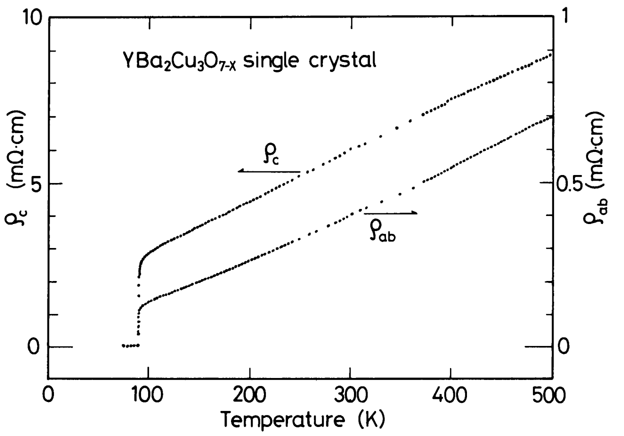


Fig. 4. An example of the temperature dependences of ρ_{ab} and ρ_c of a single crystal $YBa_2Cu_3O_{7-x}$.

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