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Field-induced electron emission from broad-area YBaCuO high-T_c electrodes

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A "transparent anode" facility has been used to compare the field-induced electron emission characteristics of planar YBaCuO high-T_C electrodes at room temperature and in the superconducting state. It was found that virgin electrodes under ambient conditions exhibit an initial "current switch-on" event at typical field levels of $\sim 30\ \mathrm{MV\,m^{-1}}$, with the emission coming from a single point site: a large hysteresis effect was also observed when the field was cycled. In contrast, at low temperatures, no switch-on events were observed, and the hysteresis effect was significantly smaller. Also, if an emission current in the nanoamp range was recorded under constant-field conditions as the temperature was lowered through T_C, there was an abrupt (i.e. step-like) fall in both the emission current and its associated noise at the superconducting transition.

1. Introduction

With the more general availability of high T_c material, there is a growing interest in its possible technological applications. Among these are device and systems applications that would require high T_c electrodes to sustain electric field strengths in excess of $10\ \mathrm{MV\,m^{-1}}$. Two specific examples, relevant to the present study, would be a superconducting r.f. cavity [1] and a high-voltage (HV) cryogenic transmission line [2]. For the more distant future, high-T_c materials could also be seen as playing a major role in space, where the necessary cryogenic conditions occur naturally; in this context, they could provide a convenient means of electromagnetic screening. An essential prerequisite for the successful (non-lossy) operation of such systems is that there is no parasitic "cold" electron emission from the electrode surfaces under operating conditions; i.e. similar to the "electron pin-holes" that frequently occur on normal vacuum-insulated HV electrodes [3]. Accordingly, there is a developmental need to evaluate the performance of these materials under high surface field stress and controlled laboratory conditions.

There is also a need to conduct a fundamental study into the physical nature of any emission process observed with these materials; in particular, to determine what influence (if any) the superconducting state has on the basic emission process. Such an investigation would therefore parallel earlier work on "conventional" superconducting metals [4, 5], which showed that the superconducting state had only a "second order" influence on the emission process.

The present investigation was a "first shot" aimed at both of the above objectives. It employed a relatively simple "transparent anode" technique to provide both spatial and temporal information about the emission processes occurring on planar high T_c cathodes. In situations where there was only one site present on an electrode, it was also possible to measure its current–voltage characteristic. The specimen stage incorporated a pumped cold-finger that was capable of cooling specimens to around 10 °C below their transition temperature: in this context, an oscillating magnet technique was used for calibration purposes. By way of a control, identical measurements were made on both different samples from the same source, and on samples from different manufacturing sources.

2. Experimental procedure

2.1. Systems

The investigation was conducted in an oil-pumped stainless steel vacuum chamber at a pressure of 10^{-5} Pa, and employed an integral purpose-designed specimen stage incorporating both the cryogenic and transparent anode systems. As shown in Fig. 1, the whole assembly is suspended vertically from the top flange of the chamber, whilst side ports are used for electrical feed-throughs and optical inspection by video camera. The high-T_c specimens were centimetre-sized discs, 3 mm thick, and were mounted on a copper stub that was in intimate contact with an externally pumped liquid-nitrogen reservoir. The temperature of the specimen was measured by a chromel–alumel thermocouple that was in contact with its base. By operating the liquid-nitrogen reservoir at atmospheric pressure, the lowest specimen

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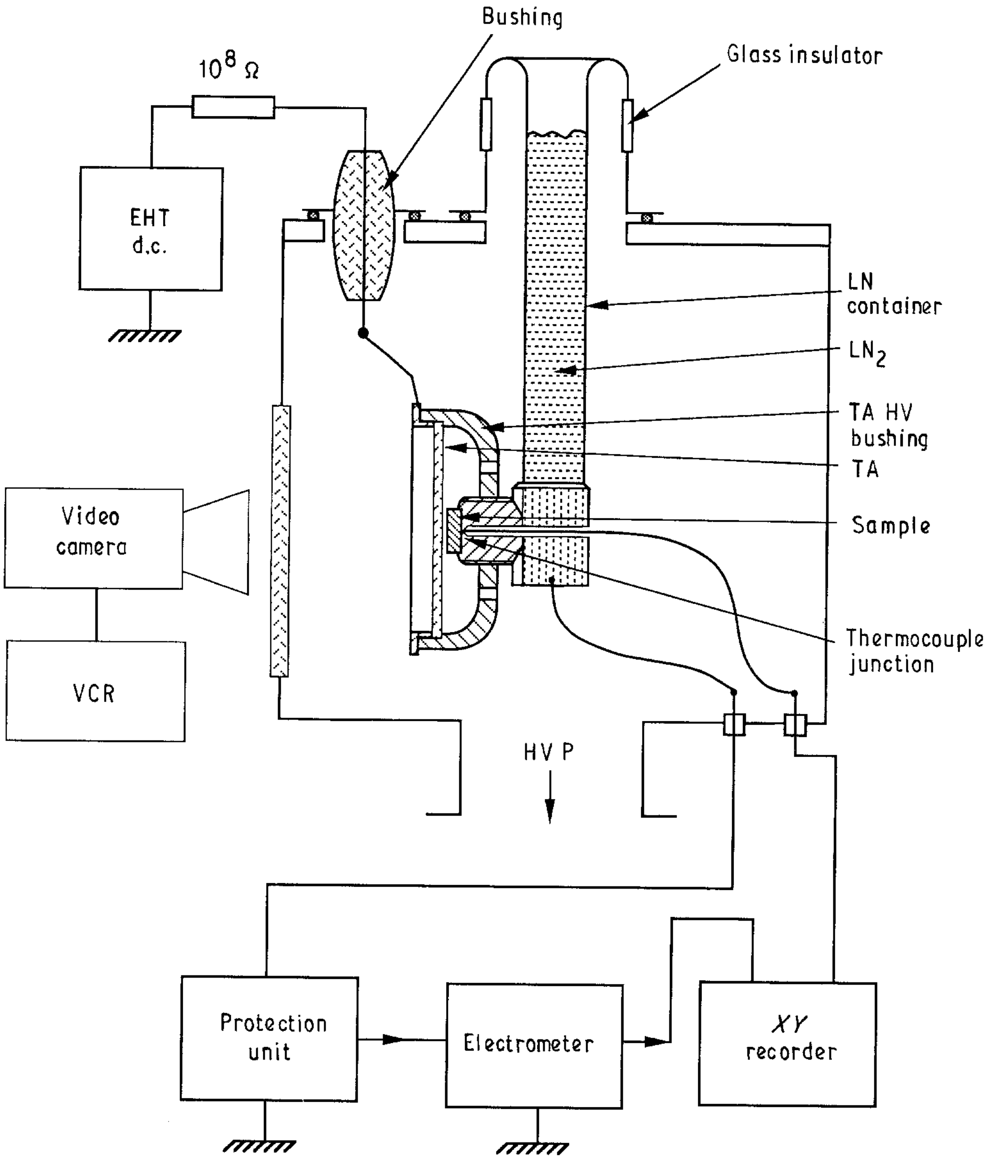

Figure 1 A schematic illustration of the experimental apparatus for the comparative study of field electron emission characteristics of a broad-area high-T_c superconductor at room temperature and in the superconducting state.

temperature attained was 81 K; however, by externally pumping the reservoir, it was possible to achieve specimen temperatures of 68 K which, as will be discussed, is well below T_c.

The "transparent anode" consists of a glass disc whose surface is made electrically conducting by coating it with a thermally diffused tin oxide layer [6]. In the present arrangement, the disc is mechanically supported by a Teflon bushing which ensures that the surfaces of the disc and specimen are parallel, and with a fixed separation, *d*. For the measurements reported in this paper, *d* was set to a room-temperature value of 0.4 mm. However, because of thermal contraction effects, this value had to be corrected at low-temperatures; at 80 K, for example, $d = 0.15$ mm. The electrical circuits used for measuring the temperature of the specimen, and its current–voltage (I–V) characteristic, are perfectly conventional and are also shown in Fig. 1.

2.2. Procedure

Because all specimens were provided in suitably sized discs, there was no need for any initial machining. Accordingly, it was only necessary to rinse ultrasonically the high-T_c specimens in alcohol prior to attaching them with silver paste to the copper boss at the base of the cold finger. At this stage, it was necessary to perform a preliminary calibration experiment to determine the T_c of the specimen; i.e. prior to applying any field to the specimen. This involved suspending a small permanent magnet in the vicinity of the specimen, and noting the temperature at which its residual oscillations are damped out, and the magnet mechanically expelled away from the sample surface [7]. Although simple in concept, this technique proved to be surprisingly sensitive, and was able to fix T_c to within $\pm$ 1 °C.

After removing the magnet, an ultrasonically cleaned anode was assembled on to the module under clean conditions, and the complete specimen stage was then introduced into the experimental chamber without delay, and immediately put under vacuum. These precautions were taken to minimize the incidence of particulate contamination of the specimen surface, because this is known to promote parasitic field-induced electron emission [8, 9].

The system was then ready for recording either the high- or low-temperature I–V emission characteristic of the high-T_c specimen, and the corresponding spatial location/distribution of any emission current. In choosing the sequence of measurements, special regard had to be paid to preserving the microscopic state of the high-T_c specimen. Accordingly, it was decided to standardize on the procedure of initiating measurements on a "virgin" specimen by first studying the low-temperature characteristic; i.e. avoiding the possibility of ambient current pulses, or microdischarges, giving rise to local damage of the electrode surface [10].

Having lowered the temperature of a virgin specimen to below T_c, a gradually increasing voltage was applied to transparent anode until a finite emission current of ~ 100 pA was recorded by the series picoammeter. At this stage, the voltage was progressively raised in small incremental steps, with corresponding current readings being recorded until its value reached ~ 0.1 μA. The voltage was then incrementally reduced in order to record the "down" I–V characteristic. For consistency, this cycle of measurements was generally repeated a number of times. The specimen was then allowed to warm up to room temperature, and the same procedure repeated. Concurrently with these measurements, a video camera (see Fig. 1) recorded all optical activity that occurred on the transparent anode. As a control, the above experimental sequence was repeated with a "commercially" polished copper specimen [11].

In an attempt to observe dynamically the normal-to-superconducting transition, a second experiment was performed, whereby a pen recorder was used to monitor the emission current under constant field conditions as the temperature was progressively lowered through the previously established value of T_c.

3. Results

The typical low- and high-temperature I–V characteristics obtained from all high-T_c specimens, and their derived Fowler–Nordheim (F–N) plots, are shown, respectively, in Figs 2 and 3. Also shown, as inserts to Fig. 2, are schematic drawings of the transparent anode images recorded at the high-current end of the characteristic. In fact, as had been noted in other studies [6], optical images of emission sites only became visible when their emission currents exceeded a threshold value of $\sim 10^{-7}$ A. Thus, when taking a specimen through the hysteresis cycle illustrated in Fig. 2, it was found that the sites "appear" and "disappear" at different fields on the "up" and "down" branches of a characteristic. From a comparison of typical pairs of high- and low-temperature images, such as those in Fig. 2, it was found that in both cases the measured current originated from the same single-point emission site. This is the first important finding, because it indicates that the "cold" emission obtained from high-T_c specimens appears to originate from the same type of localized mechanism observed with metal cathodes [3].

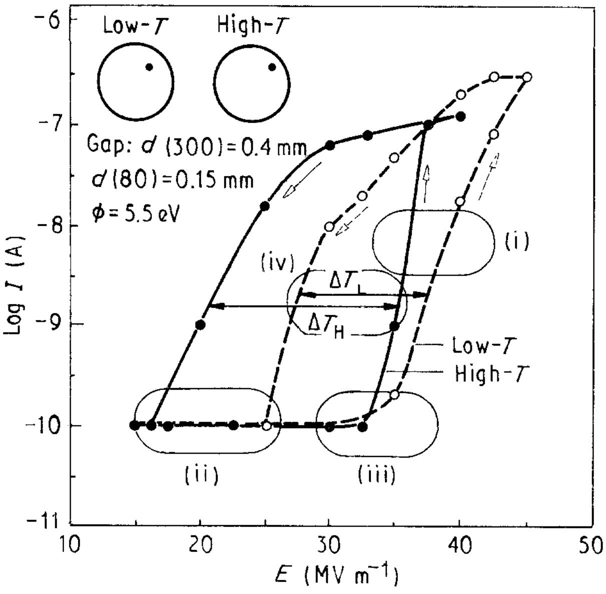

Figure 2 Typical I–V characteristics of field electron emission sites in a superconductor cathode surface at room temperature and in the superconducting state: the inserts show that the same emission site switches on at these two temperatures, as observed with the transparent anode imaging technique [6].

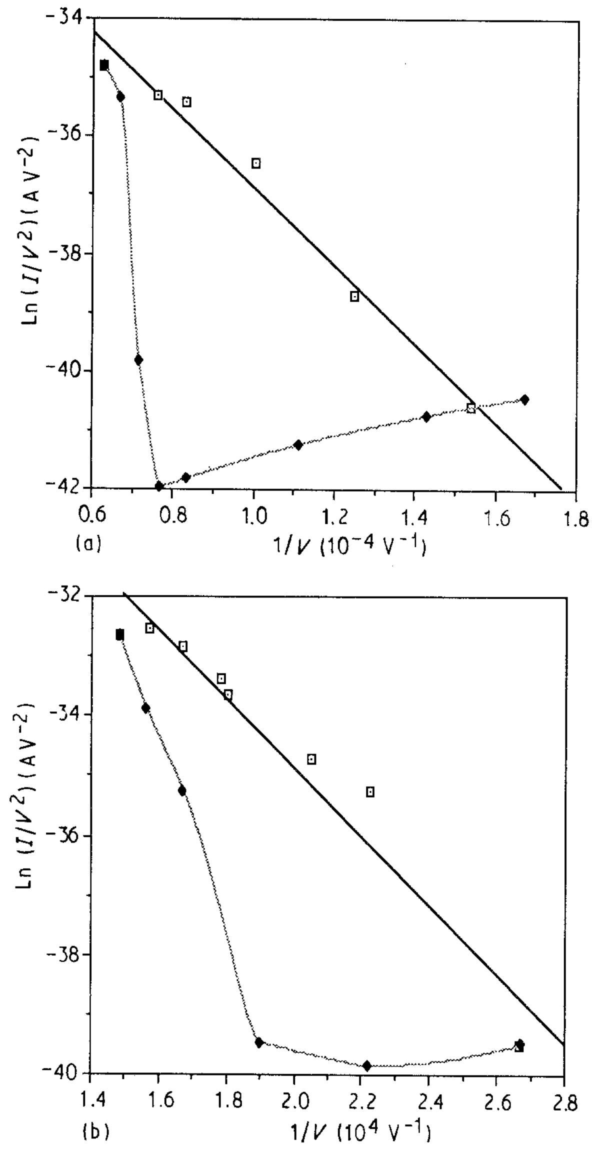

Figure 3 The Fowler–Nordheim plots of the I–V characteristics recorded (a) at room temperature and (b) in the superconducting state (□) "Down" characteristic, (◆) "up" characteristic.

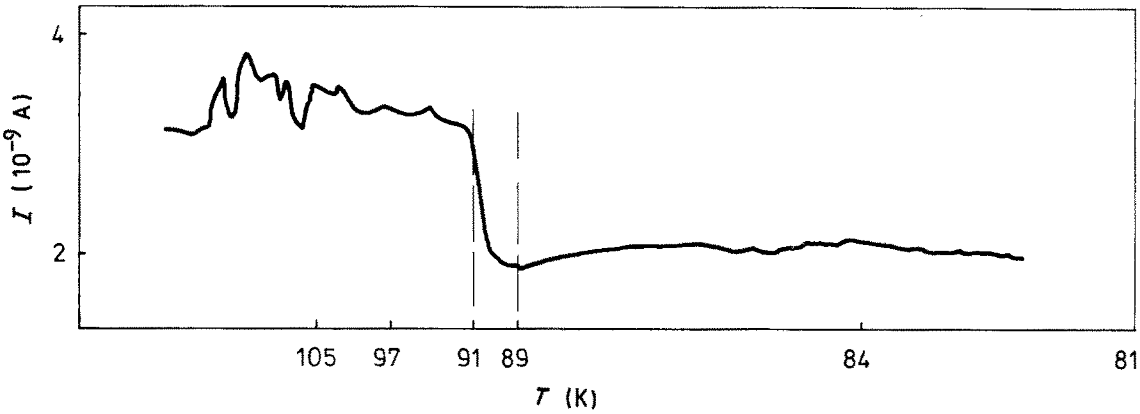

Figure 4 A typical trace showing an abrupt (i.e. step-like) fall in both emission current in the nanoamp range and its associated noise at the superconducting transition.

From an inspection of the $I-V$ characteristics, it will immediately be noted that a marked hysteresis effect is observed at both high and low temperatures. However, it is possible to identify the following four distinct differences between the low- and high-temperature behaviour. (i) The rate of current rise during the "up" branch of the characteristic is significantly greater for the high-temperature measurement. (ii) At high field levels, both characteristics exhibit an approximately linear conductivity, with the low-temperature specimen having the higher value. (iii) A broader hysteresis effect at high temperature, with a lower field threshold for current extinction. (iv) A lower field threshold for the low-temperature "up" characteristic. In addition to this "typical" behaviour, it was occasionally noted that, if successive cycles of the field were implemented over a short time scale ($\sim$5 min), the hysteresis effect vanished; i.e. once switched-on, the $I-V$ characteristic follows the "down" branch.

A particularly important observation is presented in Fig. 4. This shows how there is an abrupt transition in the signal-to-noise ratio of the emission current, recorded under constant field conditions, as the temperature is lowered through T_c. Of particular significance is the observation that this effect is only observed at very low emission currents, i.e. typically less than a few nanoamps. Furthermore, it was found that the effect disappeared after a specimen had been kept for an extended period in a vacuum environment.

Finally, Fig. 5 shows the typical behaviour observed with a control copper specimen. As will be seen, this exhibits a very similar type of behaviour to the high-T_c specimens, except that the slopes of both the "up" and "down" branches of its characteristic are steeper.

4. Discussion

In order to provide a physical interpretation of the experimental findings presented in Figs 2–5, it will firstly be necessary to summarize briefly those material properties of high-T_c materials that have a potential bearing on their behaviour under the influence of strong electric fields. Secondly, it will be necessary to outline the essential features of the localized "cold" emission process that occurs on conventional metallic electrodes.

4.1. Surface properties of high-T_c materials

The high-T_c samples used for this investigation were $YBa_2Cu_3O_{7-x}$ ceramics prepared by standard technological procedures [12] using chemically pure components Y_2O_3, Ba_2CO_3 and CuO. Bulk samples had a polycrystalline grain structure, typical of ceramic materials, and had a transition temperature $T_c = 91$ K and $\Delta T = 2$ K. The contacts between crystallites, the so-called "weak links", are responsible for limiting the critical current of the sample. Further consequences of this structural characteristic are that the material is (i) porous, with a relatively rough surface having sharp microprotrusions, (ii) fragile, with a propensity to crack during cooling cycles, and (iii) capable of storing large amounts of gas that can be desorbed in vacuum.

Of particular significance to the present study, is the important role of oxygen in the YBaCuO cell structure in strongly influencing the critical parameters of this type of superconducting material. In particular, oxygen depletion results in superconducting material being transferred into a semiconducting or dielectric state. Thus, keeping such a high-T_c specimen under

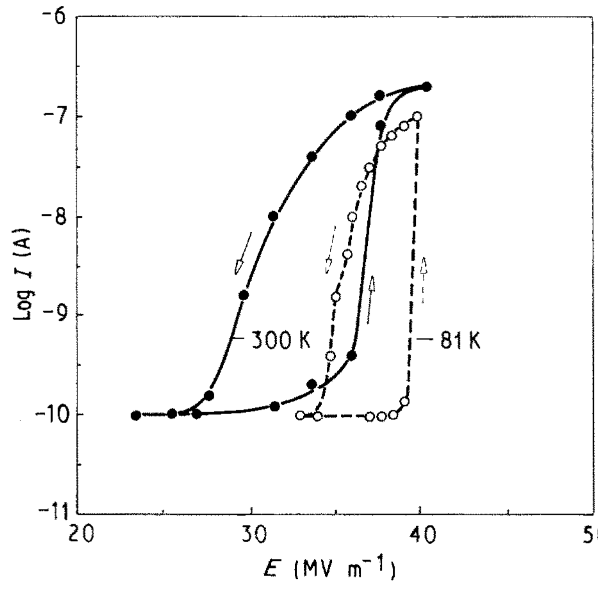

Figure 5 Typical $I-V$ characteristics of field electron emission sites in a copper cathode surface at room temperature and at a low temperature, 80 K.

vacuum conditions for a prolonged period can be expected to cause the diffusion of oxygen from the surface layers. Furthermore, this process is likely to be enhanced by the presence of a high surface electric field.

4.2. "Cold" emission from surface microstructures

From extensive studies of the "cold" electron emission process that occurs at highly localized sites on extended high-voltage electrodes at field levels in the range 5–20 MV m^{-1}, it has been concluded that the emission is almost invariably associated with the presence of micrometre-sized particulate contamination on the electrode surface [8, 9]. Thus, referring to Fig. 6, it is believed that the particle is electrically conducting, but is initially electrically insulated from the metal substrate by a "blocking" contact formed by a dielectric-like ambient oxide layer. As a result, when an electric field is applied to the surface of the electrode, it acts like an antenna and induces an enhanced field across the dielectric interface layer in the vicinity of the contact zone. Eventually, this enhanced field reaches the threshold value for inducing a dielectric switching process that creates a conducting channel and the resulting transport of electrons through the interface region [8, 13]. Finally, if this channel is located below the edge of the particle, as illustrated in Fig. 6, it is possible for some of these electrons to escape into the vacuum by a coherent scattering mechanism, and so maintain a dynamic potential difference between the particle and substrate [8, 13].

An implicit property of this model, that is particularly relevant to the present study, is its ability to predict the existence of an hysteresis effect associated with successive cycling of the I–V emission characteristic. Thus, if the surface charge associated with an emitting channel is able to leak away, or be neutralized, between field cycles, it will be necessary to apply a higher field to the next "switch-on" channel [8, 13]. Under good vacuum conditions ($\sim 10^{-9}$ mbar) with metal electrodes, such leakage is rarely observed, and one only observes an initial switch-on process with virgin electrodes. However, under poorer vacuum conditions ($\sim 10^{-6}$ mbar), where there is an abundance of residual gas ions, it is generally necessary to re-establish the charge, and so it is typical for the same metal electrode to exhibit an hysteresis effect. With the present high-T_c specimens, it was very much more typical to observe this latter type of hysteresis behaviour, although, as reported above, the former "reversible" mode was occasionally observed.

As a final, but important, general point, it can be concluded from the similarity between the behaviour of the high-T_c and copper specimens (c.f. Figs 2 and 5), that the experimental findings reported in this paper are predominantly the effect of temperature, rather than the superconducting nature of the cathode. This conclusion is supported by the work of Yankelevitch *et al.* [14] and Cabourne and Williams [15]. Yankelevitch *et al.* [14] investigated the I–V characteristics of an MIM cathode system at 300, 77 and 4.2 K, and showed that the highest conduction currents through the insulating medium occurred at room temperature, and the lowest at 4.2 K. Cabourne and Williams [15] presented field-emission data from superconducting and normal metals in the temperature range 4.2–300 K, and concluded that there was no observable change in work function when the material changed from superconducting to the normal state. They also observed discontinuities in their F–N plots, i.e. similar to that presented in our Fig. 3, but found that these disappeared with spark conditioning; an effect that was explained in terms of the removal of a surface oxide layer.

4.3. Interpretation of high-T_c emission characteristics

Because no special precautions were taken with the present high-T_c specimens to avoid contamination, it is assumed that the necessary microstructures would be present on their surfaces to support the same mechanism illustrated in Fig. 6. Indeed, this contention is further supported by the evident similarity in behaviour between the high-T_c and copper specimens (c.f. Figs 2 and 5). This model will therefore be used as a basis for our discussion.

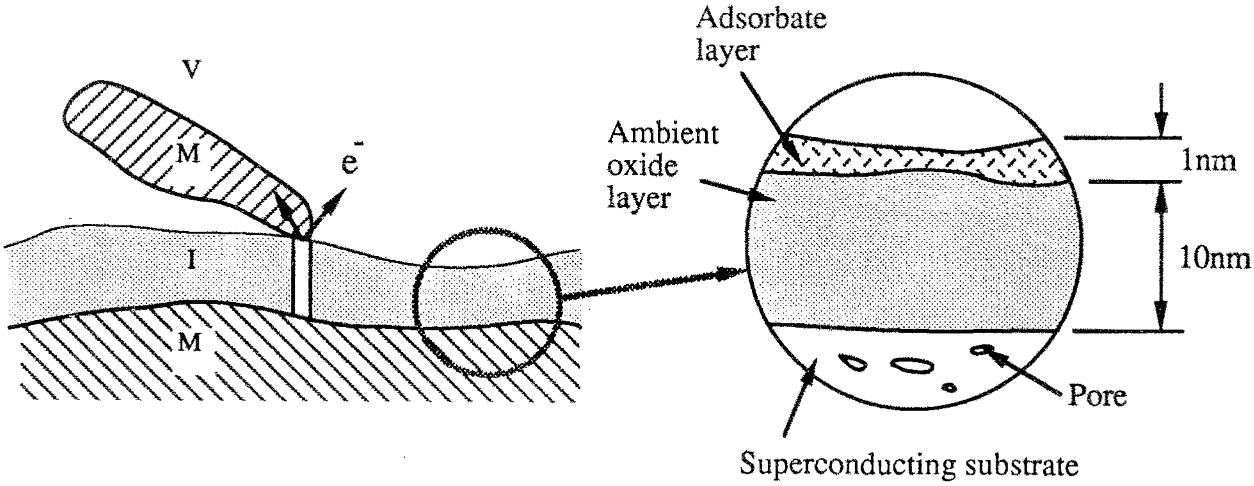

Figure 6 A schematic representation of the metal–insulator–metal (MIM) emission regime.

4.3.1. I-V characteristics

Referring to Fig. 2, it will be seen that four distinctive features have been identified for detailed discussion.

(i) Two processes can be considered that would contribute to the steeper slope of the high-temperature "up" branch of the characteristics. Firstly, and from a solid-state perspective, it can be assumed that the occupancy of the traps existing in the bulk of the conducting channel [3] will be lower at the higher temperatures, so that the channel will have a higher conductivity under these conditions. Consequently, for a given gap voltage, there will be a higher field at the metal–insulator (M–I) interface, and a correspondingly higher emission current. In addition, the lower occupancy of the traps would result in a smaller screening effect of the sensitive M–I interface and a consequently higher tunnelling current.

Because the present investigation was conducted in a relatively poor vacuum of $\sim 10^{-6}$ mPa., it is also important to consider the experimental data from an environmental perspective. In the present context therefore, it is reasonable to assume there will be a significant population of positive ions created by electron impact in the vicinity of an emitting channel [16]. These ions form a positive space charge that not only enhances the local field acting at the tip of the emitting channel, but also drastically reduces the ionisation region. The effect of this latter process is to terminate the creation of ions. It follows from this picture that the electron current to the anode is limited by the positive space charge screening. However, the external field eventually sweeps away both the remaining electrons and positive ions forming the space charge, so that the necessary conditions are restored for the cycle to be repeated under constant field conditions [18].

In this context, it should be added that the steeper slope observed with the control copper specimen probably reflects the differing surface and properties of the substrate cathode, with the most likely explanation being in terms of a more ready supply of electrons in the case of a metal cathode.

(ii) The larger hysteresis effect observed at room temperature, and particularly the lower cut-off field of the "down" characteristic, can also be interpreted as further manifestations of the effects discussed in (i) above. Thus, once a channel is in a conducting state, the lower trap occupancy and positive space charge will tend to maintain a current to lower field levels.

(iii) The lower threshold field required for observing a finite emission current under low-temperature conditions, is thought to result from two effects. Firstly, there will be a low surface charge mobility that will assist in maintaining any previously deposited charge, and secondly, there will be very few residual gas ions under these conditions to neutralize the "historical" surface charge associated with the previously emitting channel.

(iv) The somewhat higher "saturated" current levels observed at high fields and low temperatures are difficult to explain solely in terms of the channel model used to date: in fact, one would have expected the opposite effect. Accordingly, it is tempting to speculate that this result is a further "second order" manifestation of the superconducting condition; namely, the result of an enhanced tunnelling probability of electrons across the M–I interface at the base of the channel.

4.3.2. Emission current stability

In previous papers describing the behaviour of superconducting emitters, it was predicted on the basis of BCS theory that the emission in the superconducting state should be decreased by a few per cent from that measured in the normal state [5, 18]. However, in a number of experiments with both metallic [4, 19, 20] and ceramic [18, 21, 22] superconducting materials, no significant change was observed in the emission current as the temperature passed through T_c. This discrepancy between theory and experiment was explained by taking into account a number of additional factors which could influence the emission mechanism. These included the Nottingham effect and the penetration of the electric field into the emitter for the case of metallic emitters, and the effect of oxygen losses and the presence of non-metallic inclusions in the case of ceramic superconductors [5, 18].

The experiments described in the present paper show that emission comes from single micropoint sources. This implies that the current density will be relatively high, in fact higher than the critical current density, j_c, of the superconducting material. For this reason, our current versus temperature measurements were carried out at the lowest practical field levels corresponding to emission currents of $\sim 10^{-10}$–10^{-9} A. Thus, referring to Fig. 4, it will be seen that as the temperature is lowered through T_c, the current-temperature curve exhibits a marked discontinuity characterized by a "damping" of the noise on the current signal, coupled with a fall in the magnitude of the current. Significantly, this discontinuity occurred at the same temperature identified by the previously described oscillating magnet technique as the critical temperature of the cathode. It should also be noted that this result was repeatable over 12 consecutive experimental cycles, but thereafter gradually disappeared.

At the present time, we are not ready to give a final explanation for this latter effect, except to say that all the evidence indicates that the observed event is due to a localized superconducting–normal state transition in the localized surface region containing the emitter. The discontinuity itself could be due either to the creation of Cooper pairs, as predicted elsewhere [5], or as a result of the Meissner effect. In the latter case, the damping of the current signal could be the result of an interaction between the magnetic flux associated with the noisy emission current just before the transition, and the diamagnetic state of the emitter just after the transition. Another plausible mechanism responsible for this phenomenon might be associated with the possible phase transition occurring as the superconductor cathode passes through T_c. This could result in a significant change of conditions of the interface between the superconductor and the insulating layer. After a number of experimental cycles, it is believed that there will be sufficient oxygen losses from the microscopic emission region to cause a local superconducting-to-normal transition, and the consequent disappearance of the phenomenon.

5. Conclusion

It has been demonstrated that there are reproducible differences between the field emission characteristics of high-T_c specimens as measured at room temperature and ~5–8 °C below T_c. However, there is no significant evidence to indicate that the superconducting state has a major influence on their I–V characteristic: rather, they have been shown to exhibit very similar emissive properties to a control copper specimen. It was, however, demonstrated that the superconducting state has a direct influence on the signal-to-noise ratio of the emission current; also, some evidence was obtained to suggest that it had a "second-order" effect on the supply of electrons from the substrate electrode.

The detailed nature of the I–V characteristics obtained from high-T_c specimens, including a marked hysteresis effect, has been successfully interpreted in terms of a previously developed metal–insulator–metal–vacuum model involving the formation of a conducting channel in a dielectric medium. It has also been shown how this model has to be modified to take account of the presence of a residual gas atmosphere such as existed for the experiments reported in this paper.

From a technological standpoint, the findings presented in Figs 2 and 3 indicate that, compared to standard copper electrodes, high-voltage electrodes made from high-T_c material could be expected to have a significantly higher threshold field for the switch-on of a parasitic "cold" electron emission process, and subsequent breakdown events. Accordingly, there could be a potential benefit from using this type of material where HV systems and devices have to operate under cryogenic conditions, such as could be encountered in space applications. In this latter context, it would be appropriate to recall that high-T_c material could be of particular value for electromagnetic screening applications.

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