Field emission and surface conditioning

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Abstract

Vacuum breakdown is known to be the major limitation to the obtention of high electric fields in devices like klystrons or accelerating RF cavities. As a precursor to breakdown, electronic field emission has triggered much interest in fundamental as well as in applied research. The present paper describes some recent experimental results on enhanced field emission from extended metallic surfaces. Interpretation of these results favors the rehabilitation of the old geometrical model explaining the enhancement of field emission in terms of field amplification on sharp conducting protrusions. A few plausible remedies are discussed, with special emphasis on the case of field emission from superconducting RF cavities.

I. Introduction: some known things about enhanced field emission.

When an intense electric field is applied on a metallic surface placed under vacuum, an electronic emission eventually occurs. This phenomenon, known as electron field emission, is well explained by the tunneling of electrons through the potential barrier which encloses them inside the metal. The corresponding theory, proposed by Fowler and Nordheim (ref. 1) describes correctly the field emission from well characterized, very clean metal surfaces of very small area (needles). The onset of field emission is rather steep, and occurs for a threshold field of the order of 1 GVolt/m.

The behaviour of large area electrodes appears to be completely different (ref. 2). They start to emit at much lower field values, typically a few tens of MVolts/m. Emission does not take place on the whole electrode surface, but is restricted to some micron sized sites. So far, the nature of these sites has been rather elusive. Experimentally, emission from one individual site appears to obey Fowler-Nordheim (FN) law, provided effective parameters are introduced: the effective electric field must be multiplied by an ad hoc factor β of the order of 100, and the effective area of the emitter must be taken in the range $S=10^{-17} - 10^{-12}$ m^2. These parameters find a natural explanation if one assumes that the emitting site is a sharp conducting protrusion. Early researchers failed to identify such protrusions, and this is why the protrusion model fell into disfavour. Many other mechanisms have been proposed to explain the enhanced field emission from broad area electrodes (refs. 3–5), but none of them has received a full confirmation. This motivated an experimental research program at Saclay, dedicated to a better understanding of field emission from extended metal surfaces.

We developed three dedicated facilities for the experimental study of field emission. These instruments are complementary, and cover a wide spectrum of possible experiments, investigating the microscopic structure and properties of emitters, both in DC and RF regimes. Details of the apparati can be found elsewhere (refs.6–10). The main outcome of this study will be summarized here.

II. Some recent results obtained at Saclay on enhanced field emission

II.1. Natural emitters

Using a scanning electron microscope specially equipped to enable local field emission measurements and microscopic examination of the surface (refs. 7,8,10), we confirm that all emitters coincide with previously identified surface defects. When examined with the "modified SEM" facility, emitting sites were always found to be correlated with some kind of defect. The defects seem to belong to two main categories: geometrical defects, revealing no foreign element under EDX analysis, and defects from particulate contamination.

All defects do not emit: only 5% of the defects identified by investigation with the SEM emit at field levels of the order of 100 MV/m. For naturally occurring particulate contamination, we failed to identify a clear criterium saying why a given defect becomes an emitter. In particular, no special chemical composition seems to characterize emitting and non emitting particles.

Study of these natural emitters is difficult because their emission is unstable (especially in DC regime), and because they tend to be blown away if too much field is applied.

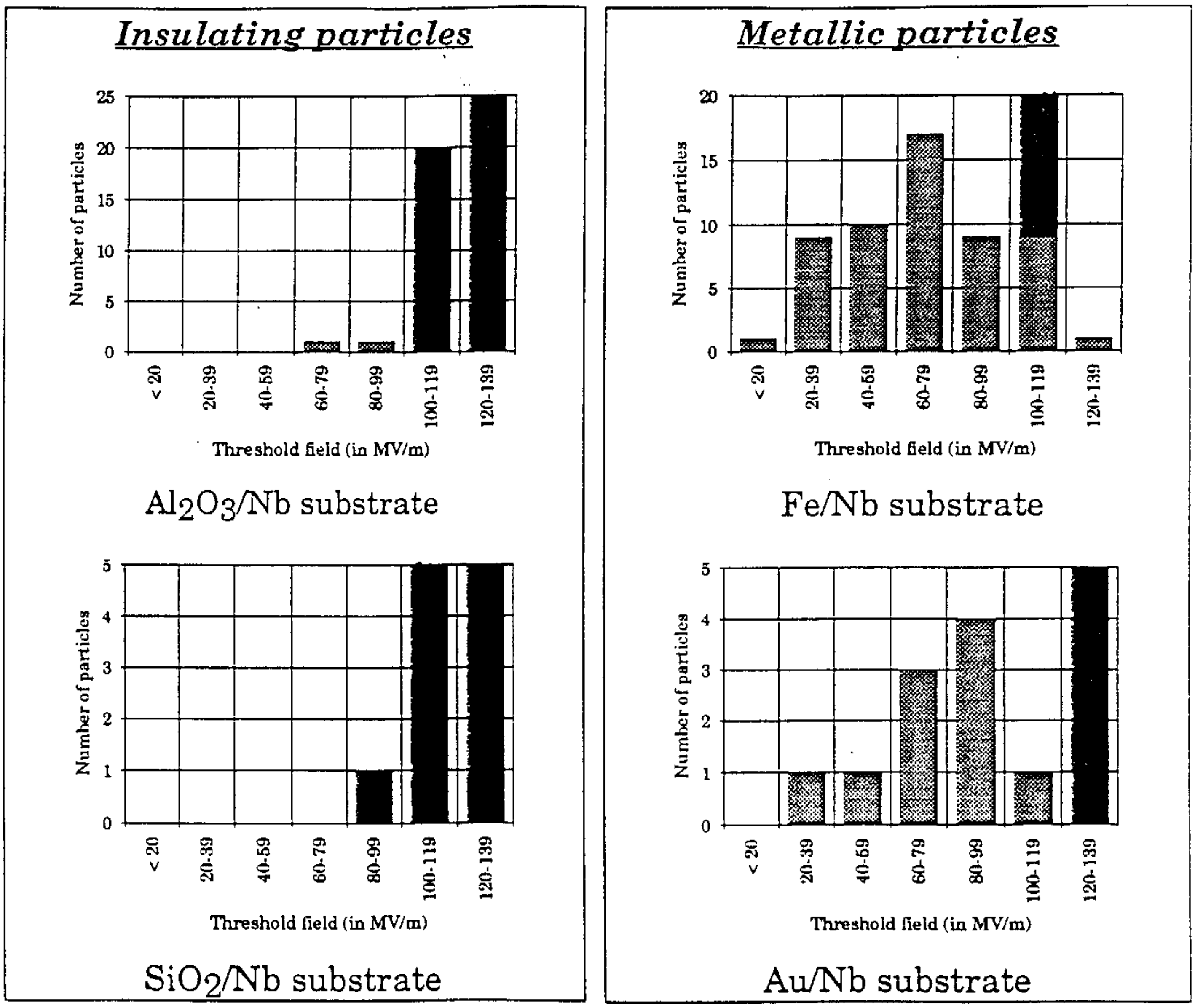
Systematic experiments have been made with a "global DC" apparatus (ref. 6) enabling the measurement of the field emission characteristics of an extended cathode in the low field, low current regime. They indicate that below 0.1 pA, the I(E) characteristics of the emission is strongly hysteretic and cannot be extrapolated from the I(E) behaviour observed at higher current. This emission is affected by adsorbed gases, and its mechanism is still unclear. It is not even clear that the emission is located at the same sites as those active in the high field, high current regime.

II.2. Artificial sites

For the reasons mentioned above (difficult study, no clear cut "zoology" of natural emitters), the forthcoming studies at Saclay were focussed on emission from well known, well controlled artificial emitters: either metallic or insulating particles of known size and morphology, or artificial geometrical defects produced by scratches.

II 2.1 Artificial particulate contamination

Powders of conducting or insulating materials were sprayed in controlled amounts on Nb and Au substrates. Very clear results emerged: most of the conducting particles (Fe, Ni, Au, Ag, Nb, Ti) behave as emitters at low field levels, whereas most insulating ones (Al_2O_3, SiO_2) do not emit, even at high fields (fig. 1).



Light bars show threshold fields for measured emission; dark bars show maximum applied field when no emission was seen.

Figure 1 Field emission threshold of insulating and conducting particles.
E_{th} is defined as the field for which the current is equal to 10 pA.

Conducting particles tend to orient themselves along the electric field lines (fig. 2). This behaviour maximizes the microscopic electric field at the particle apex. After application of the field, particles were found to be in electrical contact with the substrate, and even welded to it (fig. 3). In most cases, the welded particle still behaved as an emitter.

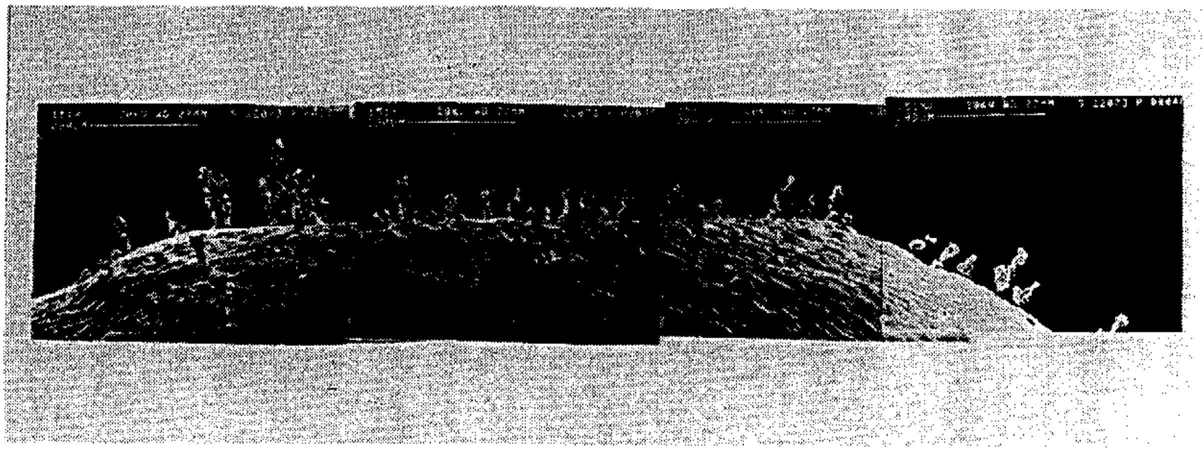


Figure 2 Orientation of iron particles along electric field lines in RF regime.

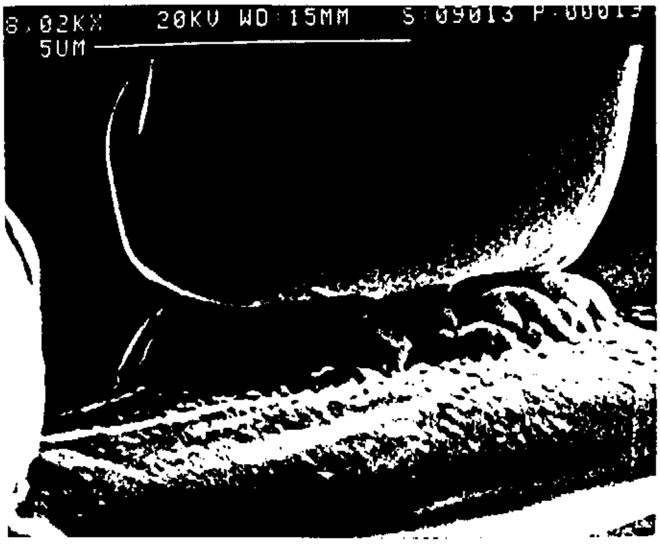
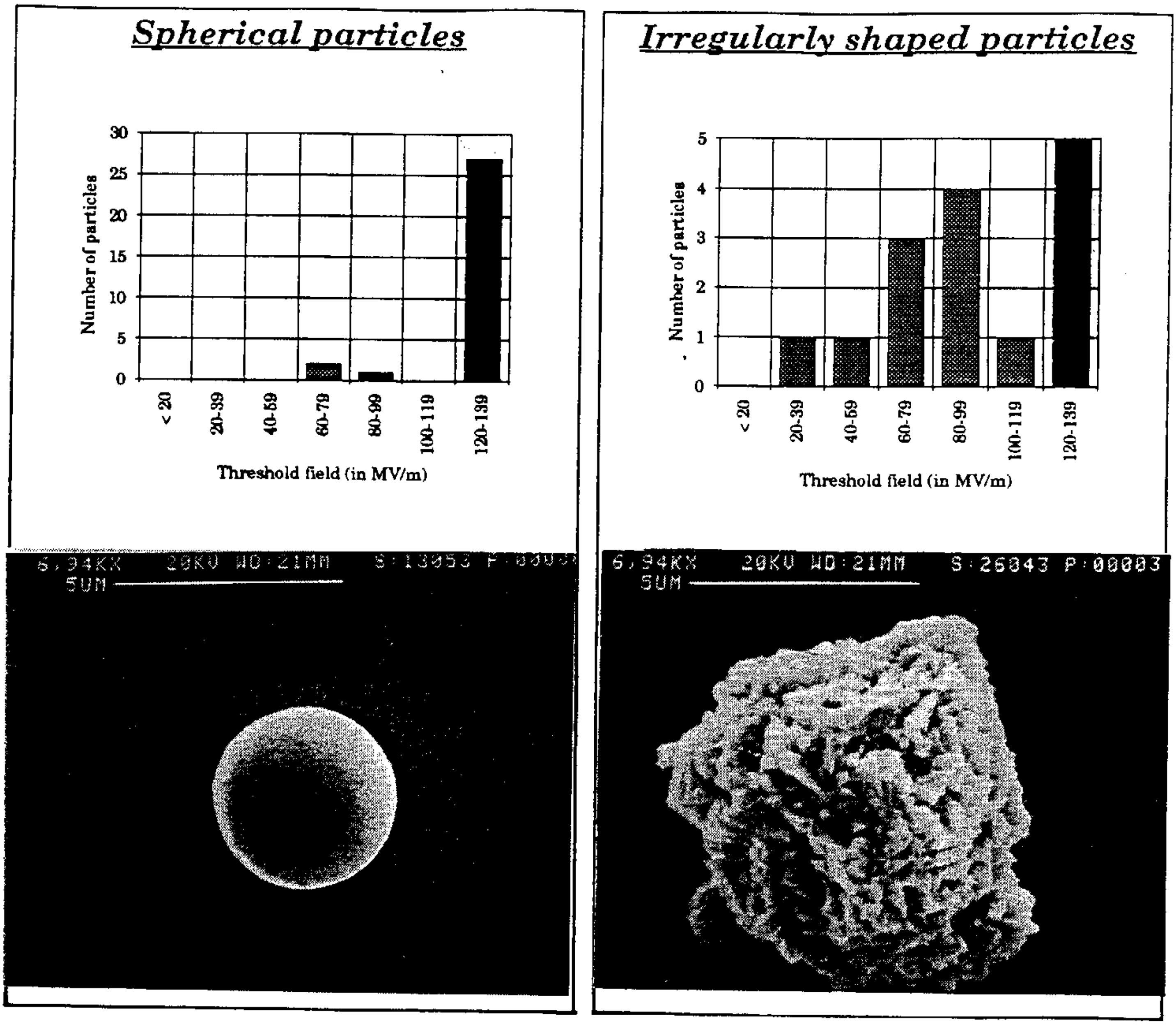


Figure 3 View of an iron particle after emission. The roll at the base of the particle is molten iron, welding the particle to the substrate.

The oxide on the substrate does not seem to play a very significant role: no difference of behaviour was observed between naturally oxidized and anodized Nb substrates, or between Au and Nb substrates.

The morphology of the particles influences emission, as is evidenced by the fact that spherical particles of Ni do not emit, whereas irregularly shaped ones emit strongly (ref. 8, fig. 4). No correlation is observed between the size and the emission of the particles.



Ni particles on Nb substrate

Light bars show threshold fields for measured emission; dark bars show maximum applied field when no emission was seen.

Figure 4 Field emission thresholds for Ni particles: a) of spherical shape; b) of irregular shape

Pulsed RF can remove dust particles. Shorter pulses seem to be more effective than long ones. This might (partly) explain the well known success of pulsed RF processing for reducing field emission in RF cavities.

II.2.2 Scratches

Geometrical defects were produced on Au and Nb substrates by scratching the surface with a needle (ref. 8). Hard needles were used (diamond, W or Nb tips). It was checked with

EDX analysis that, at least for diamond and W tips, the needle keeps its integrity during the scratching, and that no material from the needle is left on the substrate (to the precision of EDX analysis). Such geometrical defects are strong and stable electron emitters on both substrates. Morphological studies of these scratches show very sharp protrusions, which might promote a large field enhancement at their apex. Typical height of the emitting protrusions in these experiments is of the order of 10 μm. The curvature radius at the apex is commonly found to be 100 nm, and might be even less in reality, since the resolution of the SEM is of this order.

Contacts were also made with a plastic needle. Due to the difference of hardness between plastic and metal, no geometrical defects were produced on the niobium surface. No emission is observed from these contacts despite a considerable contamination of the niobium substrate by plastic particles.

II.3 For both types of emitters

II.3.1 Comparison of emission in DC and RF regimes.

Most of the studies on field emission are made in the DC regime, and it is generally assumed that the mechanisms of emission are the same in DC and in RF. This assumption has never been tested in detail, although this is clearly an important issue. The facilities developed at Saclay should enable us to draw some conclusions. Unfortunately, no clear results have been obtained yet, mainly because the comparative experiments are only beginning. Even with complete experiments, the comparison might be blurred for the following reasons: i) Adsorbed gases obviously affect field emission, and desorption is certainly more effective in RF than in DC; ii) Particulate contaminants which dominate the field emission behaviour on large area electrodes tend to be removed more easily in DC than in RF regime. It is thus difficult to ascertain that the studied sample is in the same state during measurements in DC and in RF.

Despite these difficulties, we could already check that after RF conditioning, the Fowler-Nordheim characteristics of a large area electrode is about the same in DC and in RF.

II.3.2 Thermal effects

Important thermal effects are observed on emitting sites. Like in many previous studies (ref. 11), craters of molten niobium are often found around active emitting sites (fig. 5). (more precisely : around emitting sites which *have been* active. Craters seem to appear after some discontinuity — for example a breakdown — in the behaviour of the emitter).

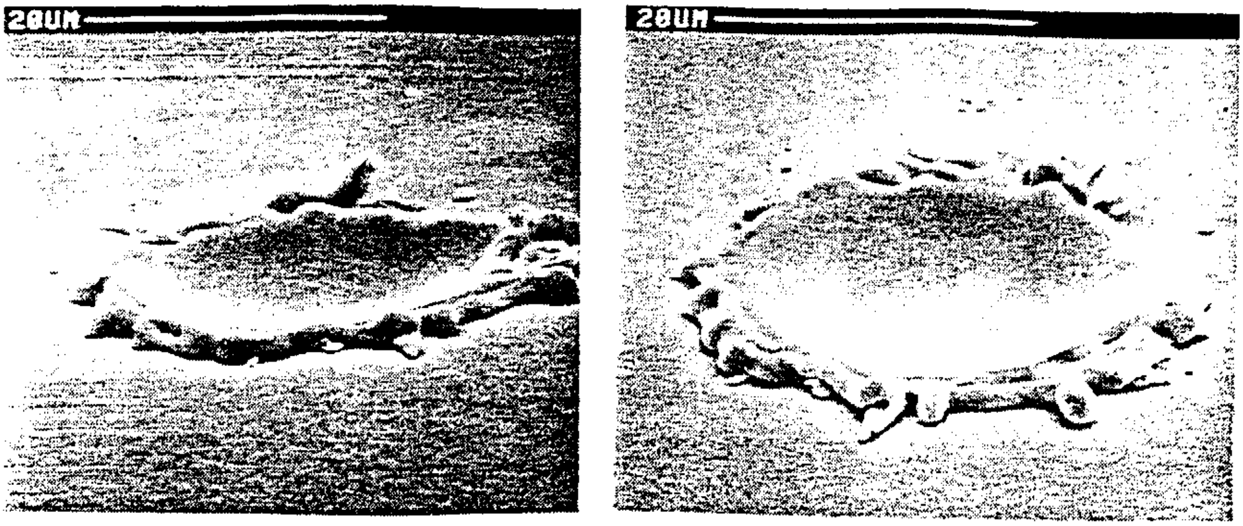


Figure 5 Typical craters (here: molten niobium). After crater formation, the sharp protrusion visible on the right hand part of the crater rim behaved as an emitter, with an emission threshold of 36 MV/m.

After emission in RF, dust particle emitters, eg Fe or Ni, appear to be molten and welded to the substrate surface (fig. 5).

"Scratch" emitters also display thermal effects : fig. 6a shows a geometrical defect acting as a powerful emitter. The characteristics of its emission changed irreversibly above some current threshold. SEM picture of the same site after this accident reveals a molten apex (fig. 6b). It is thus natural to assume that this is where the emission took place.

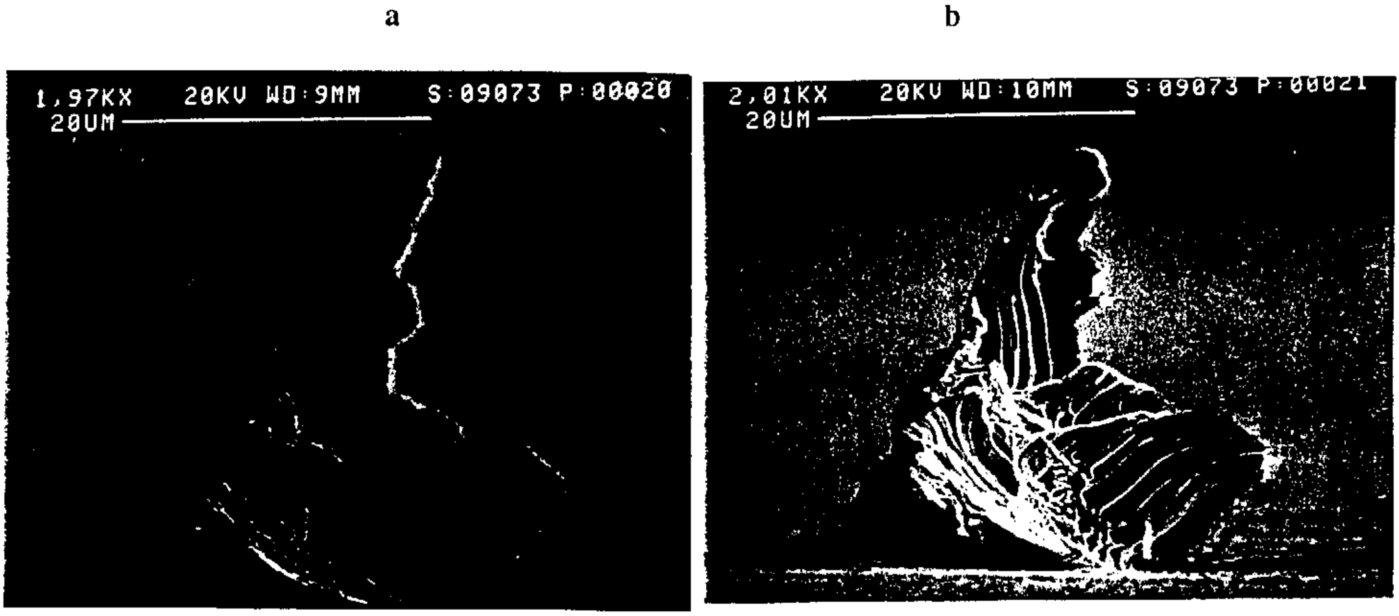


Figure 6 Morphology of a "scratch" defect (a) before and (b) after emission.

III. Discussion

III.1. Superposed protrusions ?

Most of the above mentioned experimental facts find a natural explanation if the enhanced field emission is simply due to a geometrical field enhancement at the apex of a conducting protrusion (ref. 8). This protrusion can be either a particulate contaminant, or a geometrical defect of the surface. Many electrostatic models have been published, calculating the field enhancement at the apex of variously shaped protrusions. The general outcome of these studies was that very high and sharp protrusions are needed to explain the field enhancement factors of the order of 100 required to fit experimental observations. These elusive "filaments" failed to be observed under microscopic examination, and the geometrical explanation was (too ?) soon discarded, partly for this reason. We would like to revisit this geometrical interpretation, by pointing out that field enhancements might be obtained with geometries differing from the "filamentary" ones considered so far. Evidence from many recent experiments suggests that emission might take place at the atomic scale, on aggregates of a few atoms (ref. 11). This corresponds to very sharp apex curvature radii (admitting that the concept of curvature radius keeps some meaning on this length scale), much sharper than the ones considered in the past. This can give rise to large β values, even for moderate protrusion heights. As an example, consider two superposed protrusions (fig. 7): the large one has a sharpness sufficient to produce a field enhancement factor β_1. Close to its tip, the projection surface will appear locally flat, with a uniform local field E_1 that is β_1 times greater than the field E_0 applied globally. The second, much smaller protrusion with field enhancement factor β_2 placed on its surface will itself experience a tip field E_2 enhanced over E_1 by a factor of β_2, hence an overall enhancement $\beta \approx \beta_1\beta_2$.

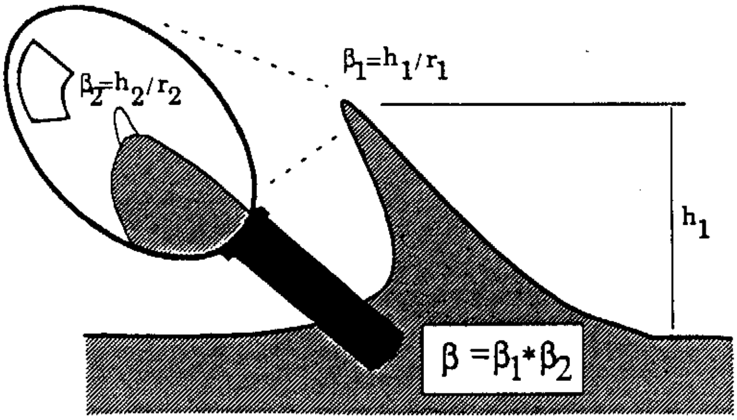


Figure 7 Superposed protrusions.

For the two considered projections, the enhancement factor $\beta_{1,2}$ equals roughly the ratio of the height of the projection to its apex curvature radius. It appears entirely plausible that a projection-on-a-projection model can account for both the observed geometry and the measured β. A beta value of the order of 100 can readily be achieved with two superposed protrusions of realistic dimensions, for example with respective heights $h_1 = 10\ \mu$m, $h_2 =$ 100 nm, and curvature radii $r_1 = 1\ \mu$m, $r_2 = 10$ nm. Such protrusions are within the range of observation of a good scanning electron microscope, and have indeed been observed by us on some geometrical defects produced by the above mentioned method. A typical example is shown in fig. 8.

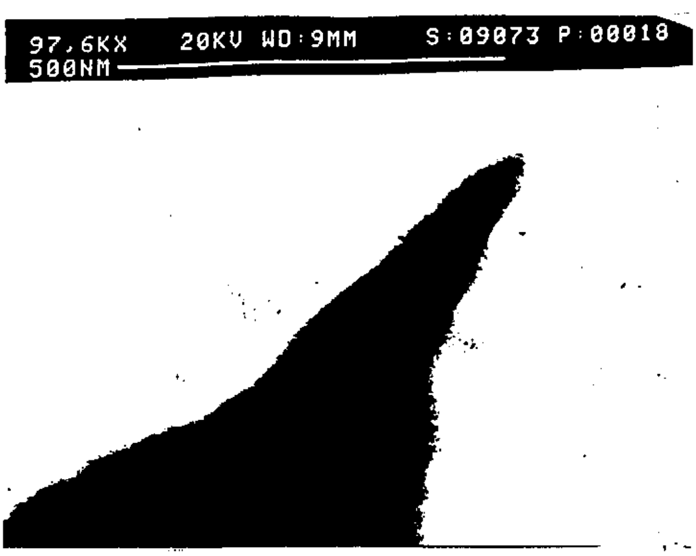


Figure 8 Same emitter as in fig. 6, seen here with a larger magnification. The apex radius is of the order of tens of nm.

All the above arguments in favor of the geometrical model must of course be taken with a large "grain of salt". We cannot ignore that field emission from protrusions is affected by adsorption or oxidation (refs. 13,14), and that these effects are not described by the simple geometrical model.

III. 2. Thermal behaviour of emitters

Obviously, field emission is not always a "cold" phenomenon. Evidence that emitters are heated during the emission process is very clear, as has been shown above. Craters are quite common in the vicinity of active emitters. In many cases, the apex of the (refractory) protrusion appears to be molten, where emission took place. Some of the metallic dust particles acting as field emitters appear to be welded to the substrate after emission. Light emission from active electron emitters has also been observed (ref. 15). In some cases, the spectrum of the emitted light was thermal.

The first of these phenomena, i.e. crater formation, is clearly related to breakdown and is time dependent. But the heat generation by the emitters, leading to apex melting, substrate welding and light emission, occurs probably in a permanent regime.

What is the origin of this heating ? The possible causes of heating have been reviewed by Swanson et al. (ref. 16). These are: i) The Joule effect due to the passage of the emission current through the narrow protrusion tip; ii) The Nottingham effect due to the energy difference of the emitted electrons and the supply electrons which replace them; iii) the bombardment of the site by ion backstream; iv) the standard heating due to the RF wave (in RF regime only).

The heat equation was solved for a protrusion of realistic dimensions, taking into account the four sources of heating mentioned above. The results of this study can be summarized as follows:

—The characteristic time of thermalization of a protrusion in good thermal contact with the substrate is very short, smaller than 10^{-7} sec. This gives a precise meaning to what we term as "permanent regime".

—The cooling of the protrusion is dominated by the heat conduction through its base; the radiation cooling plays a negligible role.

—In order to obtain an apex temperature in the 2700K range, (as is needed to melt a niobium protrusion tip), it is necessary to have a heat flux larger than 10^{11} W/m^2 at the base of the protrusion.

—In the "moderate" current range of emission studied in our experiments (I<10 μA), the Joule heating is not sufficient to explain the observed elevation of temperature. The dominant sources of heating are probably due to ion bombardment, or eventually to RF heating.

— Field emitters may be electronically and thermally unstable. Two related phenomena contribute to these instabilities: Initial heating of the protrusion can induce thermionic emission, with a drastic increase of the emission current, and further heating. Vaporization of the hot metal can generate a dense plasma with a subsequent ion backstream, also liable to enhance the heating. The relative importance and causal order of occurrence of the two mentioned phenomena probably depends whether the material is refractory or not. Breakdown and crater formation are thought to be initiated by such instabilities.

More definite conclusions about the thermal behaviour of emitters cannot be drawn because of the uncertainty on the value of some crucial parameters entering in the model, such as the protrusion apex radius, or the value of the thermal resistance between the emitter and the substrate.

Nevertheless, the present study indicates possible treatments to minimize field emission from extended surfaces: the small size of the protrusions on which field emission takes place makes them thermally and mechanically fragile. It can be envisaged to destroy them mechanically by ion bombardment. This method is used with some success in superconducting cavities, under the generic name of "helium processing". Ion bombardment with a mixture of argon and oxygen has also given interesting results on samples (ref. 17).

Thermal destruction of the emitters can also be envisaged, either by a global heat treatment of the surface, or by use of RF processing. The global heat treatment leads to a modification of the surface morphology, and has been effective on samples (refs. 18–20). Pulsed thermal treatment is also potentially interesting: with pulsed laser or electron bombardment, the very small thermal inertia of the emitter could be used to advantage. The choice of very short pulses might enable to burn the emitters, leaving the rest of the surface thermally unaffected. Apparently, this kind of pulsed heat treatment has not been tried so far.

RF processing is well known as an effective treatment to minimize field emission. It has been used successfully in many practical applications. Its efficiency probably lies in its capacity of heating selectively the emitters, initiating breakdowns which ultimately burn the emitters. As has been shown, the craters formed during the treatment may eventually emit themselves, but these emitters can be processed in turn in the same way. The efficiency of the treatment saturates when the emission from the new craters equals the emission of the formerly destroyed emitters. If this view is correct, High Peak Power Processing (refs. 21,22) can be considered as a particular pulsed thermal treatment. Fig. 9 shows an example of gradient enhancement in a superconducting cavity after such High Peak Power Processing.

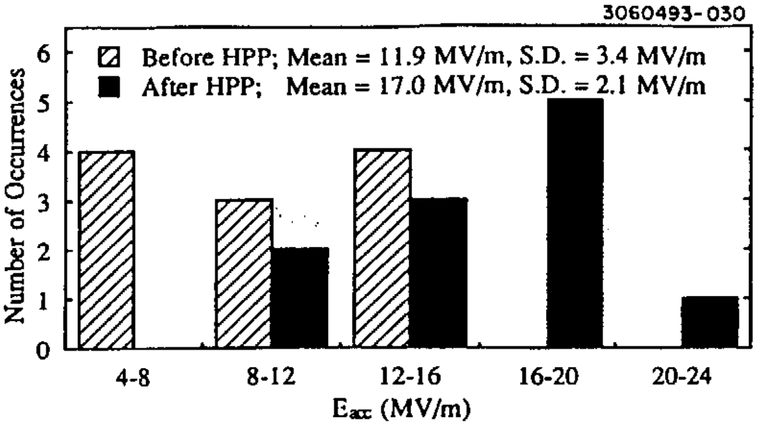


Figure 9 Gradient enhancement in superconducting RF cavities after High Peak Power Processing (from J. Graber, Thesis, Cornell University, 1993)

Unfortunately, due to the risk of ulterior contamination, the usefulness of all these treatments is severely reduced if the processed surface has to be reexposed to air. This is why *in situ* processing of the already assembled device should be preferred whenever possible. In fine, it is suggested that dust contamination is probably the most severe cause of field emission. These emitters can be treated by RF conditioning, but the efficiency of the remedy is hampered by the fact that craters and molten metal debris resulting from the processing are themselves potential emitters. The author thus feels that a rigorous cleanliness is a primary condition to avoid field emission and breakdown on metal surfaces in vacuum.

IV. Conclusion

By a series of dedicated experiments, we have found strong evidence in favor of the projection model as the main explanation for the emission of at least two kinds of sites: conducting particulate contaminants and regions of mechanical damage. It remains to be seen to what extent the sites studied here are similar to the ones actually met in practical applications. We do feel, however, that this study has some degree of generality, because all the "natural" emitters we met so far belonged to one of these two categories: conducting particles or geometrical defects. In particular, we never met evidence for more complicated emitters, like MIM structures (ref. 23).

Till now, the effort from Saclay was directed mainly towards the *understanding* of the mechanisms of enhanced field emission. Despite some progress in this direction, we do not propose any miracle remedy to cure emission. The enemy, however, has been clearly identified: we confirm that dust contamination must be avoided to minimize field emission. Geometrical defects are less of a problem: their occurrence can probably be reduced by means of appropriate chemical treatments, and by elementary precautions to avoid mechanical contacts with the surface after the treatment. Further investigations at Saclay will be directed more specifically towards the research of surface treatments reducing the activity of existing emitters. Such treatments, like firing (refs. 18–20), or high peak power processing (refs. 21,22) already exist. We feel, however, that an indispensable prerequisite to eradicate field emission is an improved surface cleanliness.

V. Acknowledgements

The present paper is merely a synthesis of a few recent experimental data on field emission, and an attempt to interpret them in a consistent scheme. The experimental effort is essentially of collective nature. The author would like to thank especially R. Noer, M. Jimenez, J. Tan, H. Safa, A. Curtoni, T. Junquera, M. Boussoukaya, A. Zeitoun-Fakiris, G. Jouve, E. Mahner, H. Padamsee, J. Graber, N. Hilleret and M. Jablonka, for providing experimental data or for useful discussions.

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