

Electron field emission from selectively contaminated cathodes

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Abstract. The electron field emission from various kinds of particles ($\sim 20 \mu\text{m}$) placed on plane cathodes has been measured in a SEM modified by the addition of a scanning anode tip. Results are similar for cathodes of Au (oxide free) and Nb (with thin insulating native oxide). In general no emission is found from insulating particles (Al_2O_3 , SiO_2) or particles with insulating oxides (Nb, Ti) under fields at least as high as 100 MV m^{-1} , while oxide free particles (Ag, Au) and those with a conducting oxide (Fe) tend to emit strongly at much lower fields. Anodizing the Nb cathode surface to $> 200 \text{ nm}$ produces no significant change in emission; after initial emission the particle/substrate contact is found to be conducting. With rare exceptions, these results are consistent with a classic metallic projection model.

1. Introduction

The study of anomalous field emission (AFE) from large-area electrodes has a long history [1, 2]. The existence of electron emission at fields some hundreds of times smaller than expected theoretically for ideal metal surfaces causes problems for a wide range of high-voltage vacuum devices, including vacuum switches, cryogenic power cables and particle accelerator cavities. AFE is known to be a localized phenomenon, resulting most commonly from particles located on or in the surface of an electrode. The mechanism(s) responsible for the emission are not clearly understood, though several theories have been proposed [3].

Most studies in recent years have focused on naturally occurring emission sites, in an attempt to relate their emission properties to those predicted by theoretical models [4]. In many cases, the origin and detailed nature of these sites have proved difficult to investigate because of their small size. Furthermore, routine observations in our laboratory and elsewhere show that only a small fraction of the contaminant particles that occur naturally on a given cathode surface are good emitters. It would be

useful to know what is special about these few particles, and whether one can avoid their presence on a given cathode.

One approach to answering these questions is to investigate the emission from different kinds of particles intentionally placed on otherwise emission-free cathodes. Studies of this kind have so far been limited to a very few contaminant materials: graphite in the form of pencil lead and Aquadag [5], pure graphite, MoS_2 , and sulphur [6]. Graphite and MoS_2 particles have been shown often to be good emitters, in contrast with sulphur particles which emit poorly. However, it has not been clear how, and to what extent, these results can be generalized to other materials.

In this paper we present the results of a study of a broader range of materials. Our initial goal in undertaking the study was to discover which, among the particle contaminants likely to be found in a laboratory environment, are most likely to produce unwanted emission. At the same time we hoped that such a study might help to clarify the mechanism(s) responsible for anomalous emission. Interesting and useful results were obtained in each respect. We find that metallic particles with no, or relatively high conductivity, surface oxides are particularly to be avoided if one wants to minimize field emission. Unexpectedly, we also find in many cases good evidence for the previously discredited 'projection' model for anomalous emission.

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2. Apparatus and procedures

2.1. Scanning field emission microscope

Our apparatus is based on a commercial SEM (Cambridge Stereoscan 120), with an energy dispersive x-ray (EDX) detection system allowing us to identify elements with atomic numbers down to six [7]. To permit electron emission studies, a tungsten anode in the form of a truncated cone with a flat tip of diameter $\sim 80 \mu\text{m}$ is mounted in the SEM sample chamber at the end of an arm, the position of which can be changed externally in each of three directions. The anode can be retracted during SEM and EDX examination of a sample. For emission studies, the anode is placed a known distance from the sample, and the latter is displaced relative to the anode with motion feedthroughs controlled either manually or by stepping motors. The air suspension system of the SEM specimen chamber provides for good stability and vibration isolation.

A connection to an external high-voltage power supply, in series with a $200 \text{ M}\Omega$ protective resistor, allows us to apply a regulated voltage of up to 6 kV to the anode. Any current flowing between the anode and the sample (itself at essentially ground potential) is registered on a picoammeter (Keithley 485). The sensitivity and stability of the system allows us to detect emission currents greater than a few picoamperes.

The SEM chamber pressure is normally about 1×10^{-6} Torr. We have not observed any dependence of emission on pressure in the range between 5×10^{-7} and 1×10^{-5} Torr.

2.2. Sample fabrication

2.2.1. Cathodes. Our cathodes are typically 1 cm wide, 1–2 cm long, and 1 mm thick. Two materials have been used—Nb (a metal much studied in previous work) and Au (to provide an oxide-free surface). Nb electrodes are electropolished [8] to produce a flatter surface than results from a standard chemical etch treatment. They are then rinsed with water and alcohol, agitated ultrasonically, and given a final alcohol rinse. To avoid the toxic chemicals necessary for electropolishing Au, these electrodes are polished mechanically with an alumina slurry, then rinsed with water and alcohol. Any remaining alumina particles can easily be detected with the EDX system and eliminated from further consideration.

Some of the electropolished Nb electrodes are anodized [9] to produce a thick oxide coating. In general, we use a single electrode, anodized over half its surface, for comparing emission with and without a thick oxide interface.

2.2.2. Contamination. The particles used as contaminants are obtained from various sources, as described more specifically below. In general, we try to work with particles roughly $20 \mu\text{m}$ in size, though we

have found no indication that size plays an important role in the emission process.

Initial attempts at contamination by simply sprinkling particles on a clean, dry electrode met with failure: in virtually every case, the application of even a low electric field to the particle resulted in its disappearance (undoubtedly by electrostatic attraction to the nearby anode). A similar effect was earlier reported by Niedermann [10], who found it necessary to affix MoS_2 particles to a Nb substrate by heating them to 800°C . A different technique is used to hold our particles in place: the substrate is held in the cool vapour above a beaker of boiling liquid nitrogen until a thin layer of moisture is seen on its surface. The particles are sprinkled on the moist surface which then dries as it warms to ambient temperature. With this treatment, the great majority of particles resist electrostatic detachment at even our largest applied fields.

2.3. Experimental procedure

For each of a number of particles on a given substrate, our standard procedure consists of the following.

2.3.1. Particle selection. This is done according to desired size (typically $15\text{--}25 \mu\text{m}$), separation from other particles (great enough to avoid confusing its own emission with any from its neighbours), and composition (that of an intended contaminant particle, as verified by EDX).

2.3.2. Geometrical parameters. Particle height is measured with the sample rotated so its surface is nearly vertical, parallel to the SEM electron beam. The sample is then restored to its normal horizontal orientation and the anode tip is moved laterally to a position some $100 \mu\text{m}$ away from the particle. Bringing the tip carefully into contact with the substrate (as seen by the appearance of an anode–substrate current, resulting from the small stray voltage persisting on the anode), we then raise the tip a selected distance (typically $50 \mu\text{m}$, as read on the micrometer controlling the vertical positioning of the anode). This procedure gives a reproducibility of about $\pm 10\%$.

2.3.3. Emission scan. At each of a series of increasing voltages, the anode is scanned in the vicinity of the particle. If and when a clear emission current (defined as roughly 100 times above background, or several hundred picoamperes) is eventually detected, the voltage is reduced and the anode is positioned so as to maximize the current, maintaining a constant anode–substrate separation, and always ensuring that the current does not rise above the level originally detected. The minimum voltage at which a significant current (typically several picoamperes) can be detected is recorded as the threshold voltage. The voltage is then switched off, the SEM beam (which was off during the scan) is restored, and we verify that the final position of the anode is indeed above the particle.

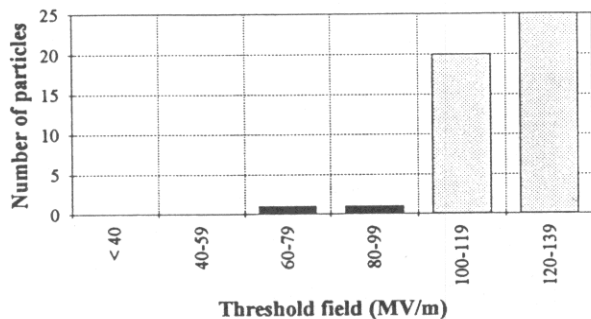


Figure 1. Emission tests for Al₂O₃ particles on a Nb substrate. Full bars show threshold fields for measured emission; shaded bars show maximum field applied when no emission was seen.

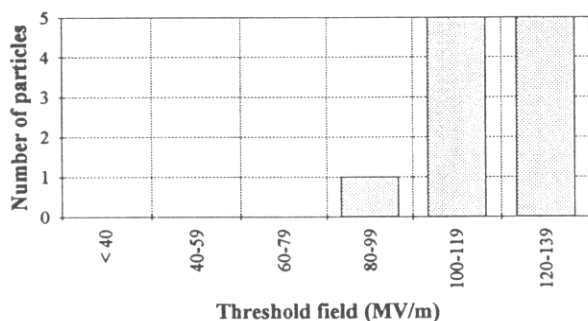


Figure 2. Emission tests for SiO₂ particles on a Nb substrate. Shaded bars show maximum field applied when no emission was seen.

2.4. Determination of fields

As we are mainly concerned with comparing emission among various kinds of particles and surfaces, precise values of the relevant threshold fields are not important. In general, we approximate the field in this case as simply $E \approx V/d$, where d is the anode/cathode separation and V the potential difference. This is a good approximation under the conditions, roughly fulfilled here, that the anode radius is greater than or equal to the separation d [11]. We ignore any effect due to the dielectric constant of an insulating particle and we ignore the local field distortion due to the shape of a conducting particle; that is, we deal only with the macroscopic field applied to the particle.

3. Results

3.1. Particles on Nb with native oxide layer

We have studied seven kinds of particles: Al₂O₃, SiO₂, Ag, Au, Fe, Nb and Ti. For discussion of their emission, they can be separated into the following three classes.

3.1.1. Insulators. Figures 1 and 2 show the results for Al₂O₃ and SiO₂. With two exceptions, to be discussed later, we see no emission below a limit of 100 MV m⁻¹. The SiO₂ came from two sources—powder (97%) obtained commercially, and crushed Pyrex glass. The latter, of course, has a significantly greater impurity content, but this appears to play no role in inducing emission.

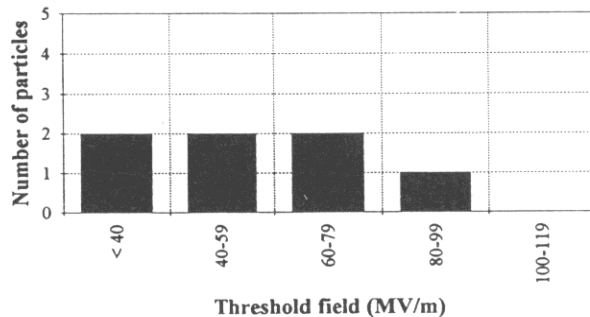


Figure 3. Emission from Au particles on a Nb substrate. Full bars show measured emission.

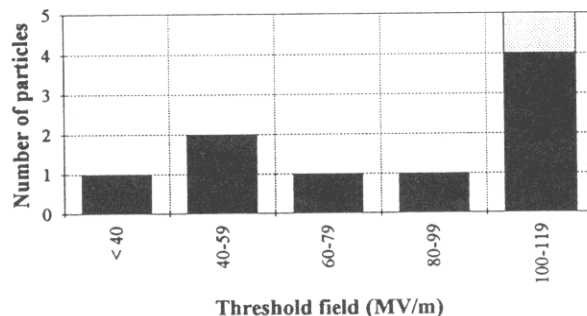


Figure 4. Emission from Fe particles on a Nb substrate. Full bars show measured emission; shaded bar shows maximum field attained when no emission was seen.

3.1.2. Metals with conducting surface. Figures 3 and 4 show the emission from Au (filed from a bar of bulk gold) and Fe (commercial powder, 99%). (A further instance of Fe can be seen later in figure 8(a).) Here we observe frequent instances of strong emission. Though considerable variation is evident from one particle to another, there appears to be no systematic difference between particles of these two elements (see discussion below). The emission from four other Au particles of considerably larger size (40–60 μm) was consistent with that shown in figure 3. We have also briefly examined Ag particles from two sources (flakes of dried silver paint and filings from a bar); their emission is similar to that for Au and Fe.

3.1.3. Metals with insulating oxide layer. As with the insulators, in the case of Nb and Ti particles (figures 5 and 6) we find no emission to at least 100 MV m⁻¹ (with one exception, to be discussed later). (Both powders were commercially obtained, with purities of 99.85% and 99.5% respectively.)

3.2. Particles on Au

To help clarify the role played by the oxide layer naturally present on Nb substrate surfaces [12], we also examined the behaviour of Fe, Au, and Nb particles on substrates of gold, where no such oxide is present. Perhaps surprisingly, the results obtained in each case were similar to those with a naturally oxidized Nb substrate. Fe and Au particles tended to emit, with a broad range of threshold fields down to 40 MV m⁻¹ and even lower, while Nb particles showed

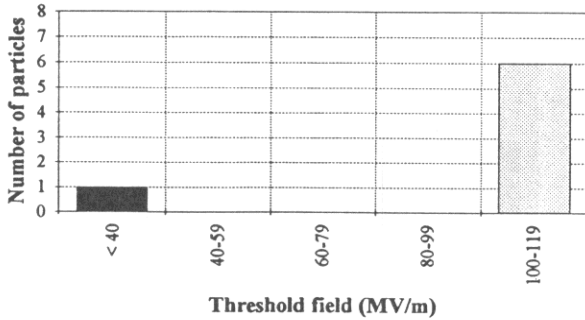


Figure 5. Emission tests for Nb particles on a Nb substrate. Full bar shows measured emission; shaded bar shows maximum field attained when no emission was seen.

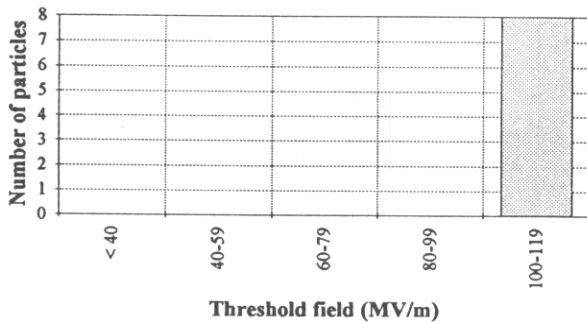


Figure 6. Emission tests for Ti particles on a Nb substrate. Shaded bar shows maximum field attained when no emission was seen.

no detectable emission (with one exception out of 17 particles measured).

3.3. Particles on anodized Nb

In a further attempt to clarify the role of an oxide layer between a metallic substrate and a metallic contamination particle, we examined the behaviour of Ag and Fe particles on anodized Nb. Figure 7 is a SEM photograph of Fe particles across the boundary between natural oxide and 240 nm of anodized oxide. The effect of the thick oxide on the contamination particles can clearly be seen in the dark halo around each particle, a well known SEM phenomenon [13] caused by the charging of the insulated particles under the impact of the electron beam; no such halo is apparent for the particles on the thin natural oxide (~ 5 nm) [12], whose conductivity is apparently sufficient to remove any charge.

Figure 8 shows the emission from Fe particles on a Nb substrate similar to that shown in figure 7. Strikingly, the presence or absence of the anodized oxide appears to make little difference. (As the emission from any given particle tends to fluctuate with time, and uncontrolled factors cause the average emission to vary from particle to particle, the small differences apparent in the figure are not statistically significant.) Figure 9 shows a similar result for Ag.

One final observation is relevant to the behaviour of contaminant particles on an anodized surface. Whereas initially all these particles charge under the SEM beam as

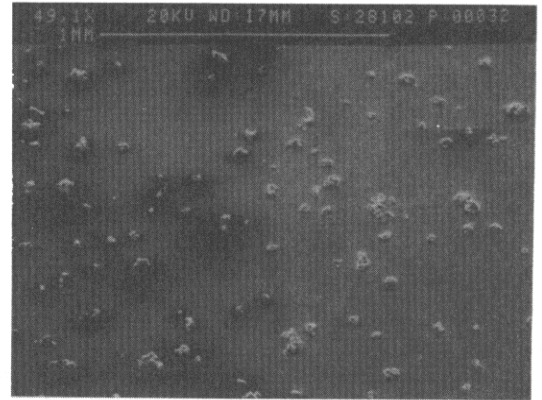


Figure 7. Fe particles on a Nb substrate. The right side of the Nb surface is naturally oxidized, while that to the left is anodized to 240 nm.

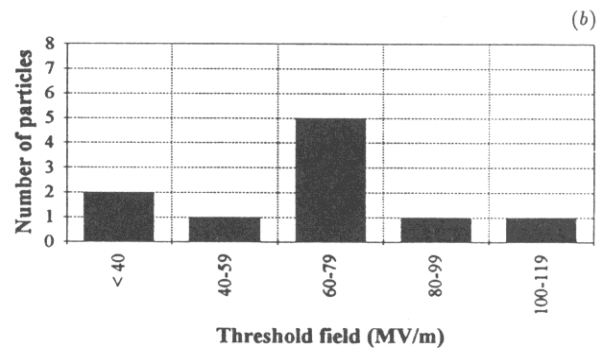
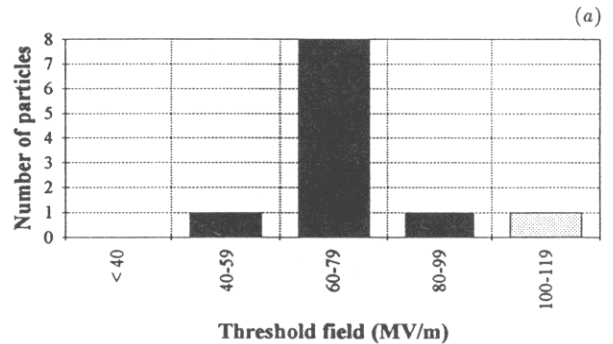


Figure 8. Emission from Fe particles placed on two regions of a single Nb substrate. (a) Nb with native oxide film, (b) Nb anodized to 240 nm oxide thickness. Full bars show measured emission; shaded bar shows maximum field attained when no emission was seen.

indicated in figure 7, after any particle emits, it loses its ability to charge. An example is shown in figure 10. The particle to the left, shown in figure 10(a) as it appeared before any application of an electric field, was then caused to emit by approaching it from the left with the high-voltage anode tip. Figure 10(b) taken subsequently shows that the particle has lost its charging halo. (The right-hand reference particle is seen to remain charging.)

4. Discussion

4.1. Theoretical models

As tentative explanations of the phenomenon of AFE, three models have been repeatedly discussed in the field

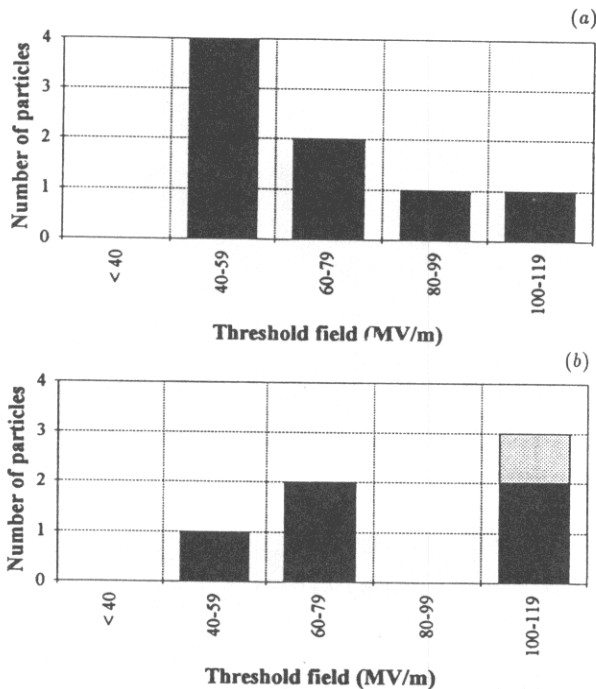


Figure 9. Emission from Ag particles placed on two regions of a single Nb substrate. (a) Nb with native oxide film, (b) Nb anodized to 270 nm oxide thickness. Full bars show measured emission; shaded bar shows maximum field attained when no emission was seen.

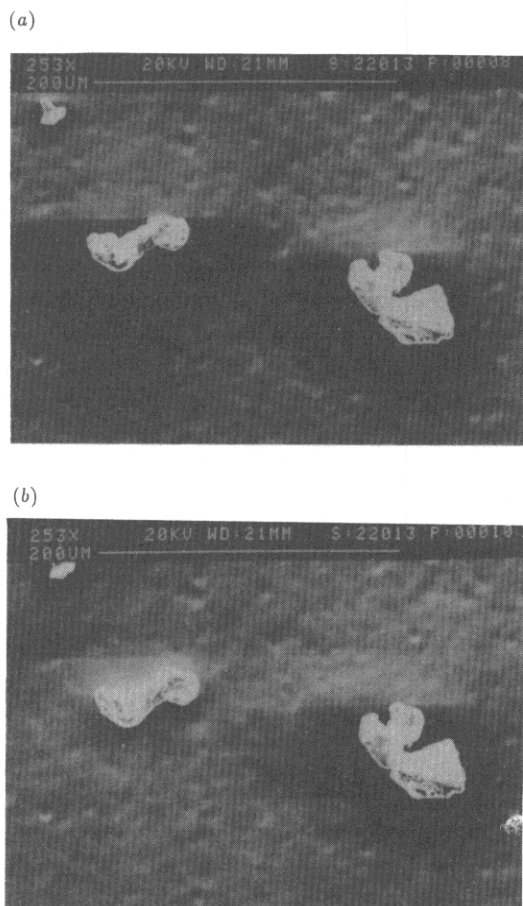


Figure 10. Particles of Fe on an anodized (270 nm) Nb surface, (a) before and (b) after application of an electric field to the left-hand particle, causing it to emit.

emission literature. The oldest, the 'projection' model [2], assumes the presence of a sharp metallic point protruding from, and in good electrical contact with, the cathode. The electrostatic distortion of the field lines in the vicinity of the point gives rise to the necessary field enhancement. This model has fallen out of favour in recent years, as observations of real emission sites with SEMs have failed to reveal projections of sufficient sharpness.

In two more recent attempts at explanation, which we refer to as the 'oxide' and 'antenna' models [4], AFE is attributed to the presence of insulating materials. The antenna model assumes an insulating layer between substrate and a superficial metal particle; the particle, acting as an antenna, serves to increase the electric field in the insulator to the point where its properties are radically altered. The oxide model assumes a comparable localized alteration in a superficial insulating particle or oxide layer (without the presence of a metallic particle) brought about directly by the application of an electric field. Both models have found support in the reported observation of insulators as well as metallic particles at emission sites, in the inevitable presence of a superficial oxide layer on many cathode materials, and in measurements of the energy spectra of emitted electrons [14].

4.2. Experimental results

The lack of emission from Al_2O_3 or SiO_2 is striking. It suggests at the very least that the special conditions required by an oxide emission model were not fulfilled to any significant degree in 55 of the 57 particles of these two insulating materials that we tested. The two exceptions, where emission thresholds of 72 and 80 MV m^{-1} were found, were among the first insulating particles we examined, before we appreciated the general trend. It is possible that these particles were misidentified, but it is also possible that they represent rare but significant instances of emission from insulating particles.

The relatively strong emission from Ag, Au and Fe particles is also clear. These metals have oxide free (Ag, Au) or conducting oxide (Fe) [15] surfaces. Though there is considerable variation in emission from particle to particle (due, no doubt, to differences in particle shapes and particle/substrate contacts), and indeed in average emission from sample to sample (due, we suspect, to differences in the moisture layer used to anchor the particles), it is clear that the striking difference in emission between these particles and the insulators we examined is real. It is also clear that the thin layer of natural oxide naturally present on Nb substrates does not play a major role, at least in our range of fields and currents, as we found little difference in the emission from similar Fe and Au particles placed on Au substrates.

On the other hand, the lack of emission observed from Nb or Ti particles shows that not all metallic particles are good emitters. It is well known that

when exposed to air Nb [12, 16] and Ti [17, 18] develop superficial insulating oxide layers, and we assume that it is this layer that inhibits emission. Furthermore, our experiments with anodized surfaces (to be discussed next) suggest that it is the oxide layer at the particle–vacuum interface (as opposed to that between particle and substrate) that plays the dominant role. (As with the insulating particles, it is not clear what importance should be attributed to the two exceptional particles (Nb on Nb, Nb on Au) that were observed to emit.)

One might expect that a thick anodized oxide layer on a Nb surface would greatly inhibit emission from strongly emitting particles (e.g. Fe). In fact, we found no significant difference in emission between particles on natural and anodized Nb substrates. (This observation is in good agreement with previous results for graphite emitters [6].) Furthermore, emitting particles on a thick oxide layer were seen to lose their ability to charge under the SEM beam. Apparently, once emission begins it destroys or renders conducting the oxide layer at the contact between particle and substrate. (To study how emission is initiated across a thick oxide layer would require the study of lower currents than our apparatus is capable of detecting.) Thus it appears that, in our range of fields and currents, any role played by an oxide barrier between a metal particle and the cathode is minimal.

Further evidence that strong emission may occur from a metallic particle placed on a cathode, without the insulating interface assumed by the antenna model, comes from two kinds of observations. The first is the good emission observed from Au particles on a Au surface, where one expects at best a very thin layer of superficial contamination. The second is provided by several instances where a particle, after emitting current in the microampere range, is found to be ‘welded’ to the surface (even as it continues to emit at low fields). Figure 11 shows a case where we succeeded in removing such a particle by pressing laterally with a tip of radius smaller than that of the particle; the particle appeared to resist the initial pressure of the tip, and, once removed, left a contact region showing evidence of melting.

Thus it appears that, at least in some cases, and in spite of recent uncertainty, anomalous field emission may indeed occur in general accord with the projection model. Nevertheless, it remains unclear how this model can account for the magnitude of the emission observed. The threshold fields measured, typically 50 MV m^{-1} , are still something like a factor of 50 smaller than those predicted for an ideal flat surface; although our particles are quite irregular, we see no features sufficiently sharp to account for such a factor. They may yet exist on a scale too small for observation with our facilities.

4.3. Implications for practical applications

The results reported in this paper have important implications for any situation where it is desired to minimize anomalous electron emission. It has long been clear that particle contaminants are a (probably *the*) major source of unwanted field emission in practical

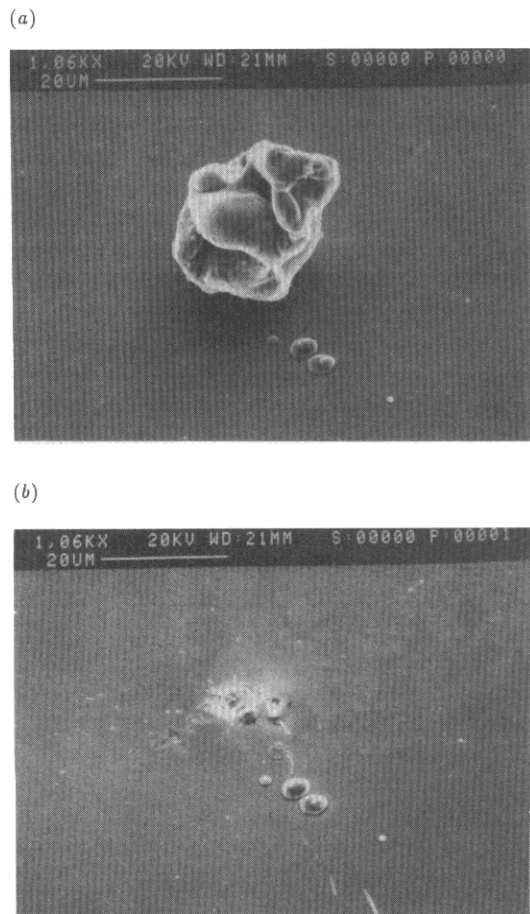


Figure 11. Before (a) and after (b) removal of a formerly emitting particle of Fe from a Nb surface.

applications. Our results show that not all types of particles are harmful. Insulators like silica and alumina, or insulating ‘dust’ in general, appear far less serious than metal particles, particularly when the latter have thin conducting oxide (e.g. Fe) or naturally oxide free (e.g. Ag) surfaces, or are produced and maintained oxide free in the vacuum environment of a high-field structure. Metal particles with conducting oxides abound in technical environments (e.g. stainless steel, or indium used for vacuum joints), and metal particles produced under vacuum might include material abraded from internal moving surfaces or Ti from sublimation pumps. A reduction in these specific kinds of contaminants could be an effective means of attacking the field emission problem.

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