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ON THE RESOLUTION AND IMAGE INTENSITY OF THE FIELD-ION MICROSCOPE

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Cleveland, Ohio

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ABSTRACT

The voltage required to produce blurring of a field-ion image spot was measured as a function of tip temperature. The results indicate that (1) the ion current causing the blurring originates at an adsorbed phase on the surface, and (2) a transit-time blurring mechanism operates at low temperatures, whereas at higher temperatures an orbit-time blurring mechanism best explains the data. The explanation of the failure of the (110) plane in tungsten to image successfully above 5 K is shown to lie in the imaging process itself, not in the action of the blurring mechanisms. Maximum resolution was obtained at 5 K, not at 21 K as generally believed. A method of image intensification is described that has advantages over other existing methods.

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SUMMARY

The voltage required to produce blurring of a field-ion image spot was measured as a function of tip temperature. The results indicate that (1) the ion current causing the blurring originates at an adsorbed phase on the surface and (2) that a transit-time blurring mechanism operates at low temperatures, whereas at higher temperatures an orbit-time blurring mechanism best explains the data. The explanation of the failure of the (110) plane in tungsten to image successfully above 5 K is shown to lie in the imaging process itself, not in the action of the blurring mechanisms. Maximum resolution was obtained at 5 K, not at 21 K as generally believed. A method of image intensification is described that has advantages over other existing methods.

INTRODUCTION

This report describes the results of several experiments performed with the field-ion microscope. The investigation resulted in information concerning the physical processes involved in the operation of the microscope. The main points discussed are (1) the resolution, (2) image intensification, and (3) low-temperature operation.

The resolution of an image spot on the screen of the field-ion microscope increases as the applied voltage is increased. There is, however, a voltage that produces maximum resolution, beyond which the resolution abruptly degenerates. A knowledge of the mechanism responsible for this sudden blurring is desirable not only because of the insight it would give to the detailed understanding of the operation of the microscope, but also because it might indicate ways of controlling the blurring, and thus lead to increased resolution.

The blurring mechanism is investigated by measuring the variation of the voltage necessary for the onset of blurring as a function of tip temperature and tip radius. The results are interpreted in terms of a two-mechanism theory: a transit-time mechanism at low temperatures and an orbit-time mechanism at higher temperatures.

There is a need for image intensification in field-ion microscopy. This is especially so for operation with small tip radii and for operation with imaging gases other than helium. A new technique for increasing the intensity of the field-ion image developed at this laboratory is also described along with a brief description of previously used methods and their limitations.

In the course of these investigations, it was observed that maximum resolution of the field-ion image occurred not at 21 K as was generally assumed (ref. 1), but at 5 K. Some comments on this observation are given in the section Low-Temperature Operation.

RESULTS AND DISCUSSION

Resolution

As the applied voltage is raised during operation of the field-ion microscope, a point of maximum resolution is reached, followed by a sudden blurring as the voltage is increased slightly above the optimum value.

As indicated in the literature, there are two possible sources for the ions producing this blurring effect: (1) the gas phase above the tip surface (ref. 2) and (2) an accommodated or partially accommodated phase on the surface (ref. 3). To distinguish between these two sources, a measurement of the voltage required for blurring onset as a function of tip temperature was made (ref. 4). Here, lack of temperature dependence would indicate an origin in the gas-phase. Conversely, if there is a dependence, then at least partial accommodation has taken place before ionization.

The microscope used in these measurements is shown in figure 1. A modified Swenson technique (ref. 5) is used to cool the tip, which is operated at ground potential. This technique consists of drawing gaseous and/or liquid helium from a reservoir through a heat exchanger in thermal contact with the tip. This method is very efficient with respect to coolant use, especially when temperatures above the coolant boiling point are to be maintained. The screen and accelerating cone are cooled by means of a liquid-nitrogen reservoir. A gold - 2.1-percent-cobalt versus copper thermocouple in the heat exchanger is used to measure the tip temperature, which, according to calculation, should be accurate to within 1 K. The background pressure is of the order of 5×10^{-9} torr.

The heat exchanger is thermally isolated from the screen-tip region. The tip wire is introduced into the screen-tip region through a small aperture. This permits the temperature of the tip to be varied, while the tip surroundings (imaging gas, accelerating cone, screen, etc.) remain at 78 K.

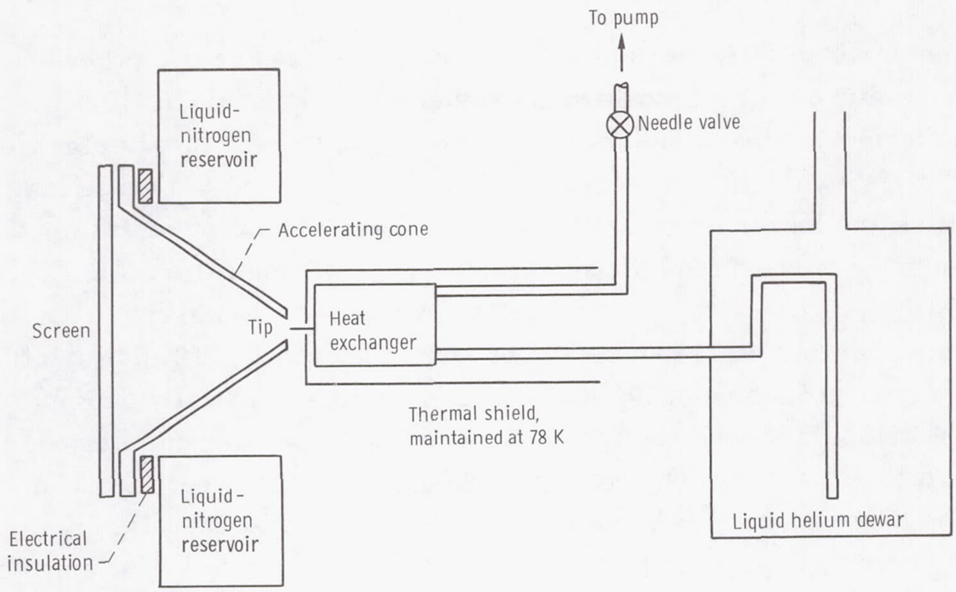


Figure 1. - Field-ion microscope.

Figure 2 shows a typical set of data for the (111) plane of tungsten with helium as the imaging gas. Additional experiments have shown that the results are independent of gas pressure, which was varied from 0.2×10^{-3} to 0.8×10^{-3} torr (gage readings, uncorrected for helium gas). The results were shown also to be independent of the temperature of the tip surroundings by warming the screen and its liquid nitrogen reservoir to room temperature, exposing the tip to about a 2π-steradian solid angle of surfaces at 300 K.

The fact that the blurring threshold is dependent on the tip temperature indicates that the blurring current originated from accommodated or partially accommodated atoms.

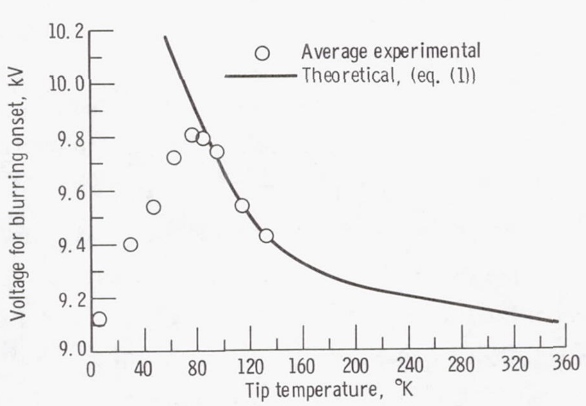


Figure 2. - Comparison of equation (1) with experimental data. Scatter in voltage measurements, about 2 percent. (In eq. (1), surface field has been set equal to 4.5 V/Å (4.5×10^{10} V/m) for applied voltage of 9775 volts.) Pressure, 0.5×10^{-3} torr (uncorrected); tip radius, 260 Å (or 10^{-10} m).

The maximum at about 80 K leads one to suspect that there are two processes operating. The low-temperature process would seem rather easy to explain qualitatively using a transit-time theory for the atoms moving in the comparatively low-field regions between the imaging tungsten atoms. A decrease in tip temperature would cause lower helium-atom velocities and longer transit times through these secondary ionization zones. This would increase the current from these areas because the ionization rate is proportional to the transit time. Therefore, a decrease in temperature would require a decrease in field to prevent between-atom ionization. A lack of knowledge of field variation in these regions, however, makes calculation of the field-temperature relation impractical.

The high-temperature region of the curve, as well as being unexpected, is a little more difficult to explain. One approach that seems to fit the experimental data consists of postulating the existence of two separate regions of ionization: (1) A region close to the surface where the equipotentials closely conform to the atomic structure of the surface and where, if under the proper conditions of field and temperature ionization does take place, the ions diverge only slightly and form a well-resolved spot on the screen. (This is the source of ions during normal operation of the microscope.) (2) A second, broader region at some distance X_{c2} from the surface, separated from the first region by a region of low ionization probability. This second region, being further from the surface, would produce a more diffuse imaging beam on the screen.

Experimental evidence for the existence of such ionization bands is given by Jason et al. (ref. 6), who have used mass spectrographic techniques to resolve structure in the energy distribution of ions produced near a tungsten tip. Their work, however, was done on the hydrogen-tungsten and the neon-tungsten systems, and it is assumed here that a similar structure exists for the helium-tungsten system.

It is postulated here that the tangential spreading of Jason's lowest energy deficit band, adjacent to the surface, with increasing field strength is the cause of the low-temperature blurring current. It is further postulated that the bands of higher energy deficit correspond to the ionization region, distant from the surface, which is the source of the high-temperature blurring current.

To be more explicit, a simple calculation may be performed to check the general fit of the model. First, it is assumed that the group of Jason's ionization regions above the first low-energy-deficit band is one broad ionization zone and that the helium atoms at higher temperatures orbit into this zone. Then, since the probability of ionization in this zone is proportional to the time spent orbiting in it, it follows that, for small ionization rates, the probability of ionization (following Gomer's derivation, p. 78 of ref. 2), is

$$c = \frac{2t_2}{\tau} = 10^{16}(2m)^{1/2} \left(\frac{r_t}{\alpha F_o^2}\right) \left[kT - 2\alpha F_o^2 \left(\frac{X_{c2}}{r_t}\right)\right]^{1/2} \exp - \left(\frac{0.68\ I^{3/2}}{F_o}\right) \tag{1}$$

where $2t_2$ is the time spent by a hopping atom in the region above the cutoff distance X_{c2} (ref. 5), m is the gas-atom mass, r_t is the tip radius, F_o is the surface field, α is the gas-atom polarizability, kT is the initial gas-atom kinetic energy, T is the tip temperature, I is the gas-atom ionization potential, and τ is the ionization time, which is approximated (ref. 1, p. 86) by

$$\tau = 10^{-16} \exp \left(\frac{0.68\ I^{3/2}}{F_o}\right) \tag{2}$$

If the condition that must be satisfied for the onset of blurring at any temperature is that c is constant, then the field-temperature relation may be calculated by adjusting c and X_{c2} to fit the experimental data. For simplicity, the variation of X_{c2} with field is ignored, and it is assumed that the field is proportional to the inverse square of the distance from the center of curvature of the tip.

Figure 2 shows the fit to the high-temperature experimental data. The agreement is good, and the theoretical curve gives reasonable values of the blurring voltage up to and beyond room temperature. The parameter X_{c2} was found to take on the reasonable value of 2.92 Å (2.92×10^{-10} m).

Next, measurements were made as the tip radius was varied to see how well equation (1) predicted the change in the voltage-temperature characteristics as the radius changed (ref. 7). Field evaporation was used to change the tip radius. Measurement of the tip radius was done by counting the rings between the (110) and the (112) planes.

Figure 3 shows the field, calculated from the applied voltage and the measured radius, versus tip temperature for several different radii. Figure 4 is a plot of T_{max}, the temperature at maximum field, against the tip radius. The data in figures 3 and 4 correspond to the same tip.

In order to calculate the theoretical T_{max}, tip radius relation from equation (1), it is necessary to know how the field maximum F_{max} varies with tip radius. This information can be obtained either from the experimental data (i.e., from the measured V_{max} and the measured radius) or by assuming an expression for the low-temperature process and calculating F_{max} from it and equation (1).

However, because the error in these radius measurements (about 5 percent) precludes using the data to obtain F_{max}, one must thus assume an expression for the low-temperature process.

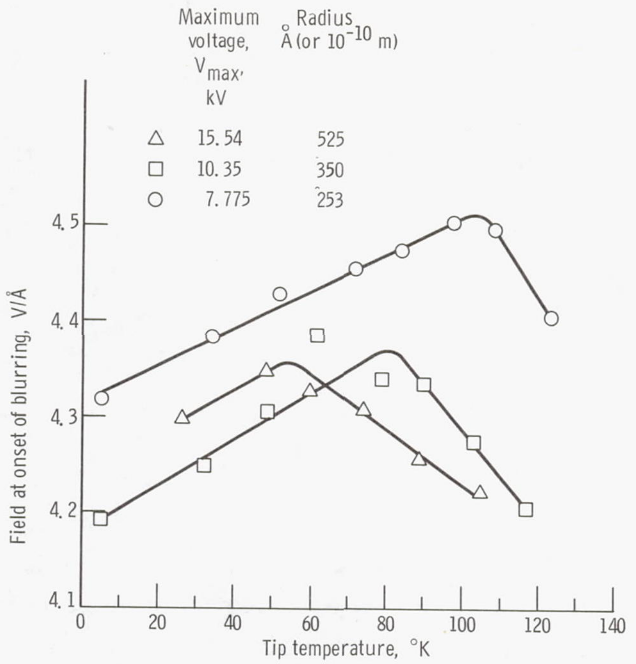


Figure 3. - Plot of field necessary for blurring onset versus tip temperature for several radii. Field calculated from applied voltage and measured radius. Because of uncertainty in radii used, calculated fields are accurate only to about 5 percent.

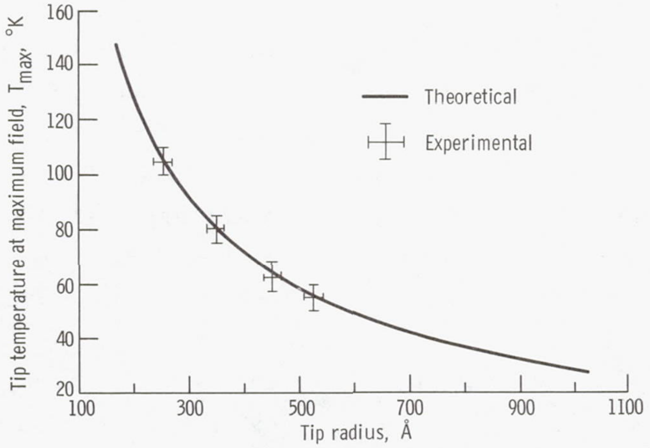


Figure 4. - Variation of tip temperature at maximum field with tip radius; comparison of theory with experiment. In calculations, surface field was assumed to be 4.355 volts per Å (4.355×10^{10} V/m) for applied voltage of 15 540 volts and tip radius of 525 Å (525×10^{-10} m).

The assumptions made in this regard are (1) that the low-temperature field-temperature curve is linear in the range of interest and (2) that the low-temperature expression is independent of the tip radius.

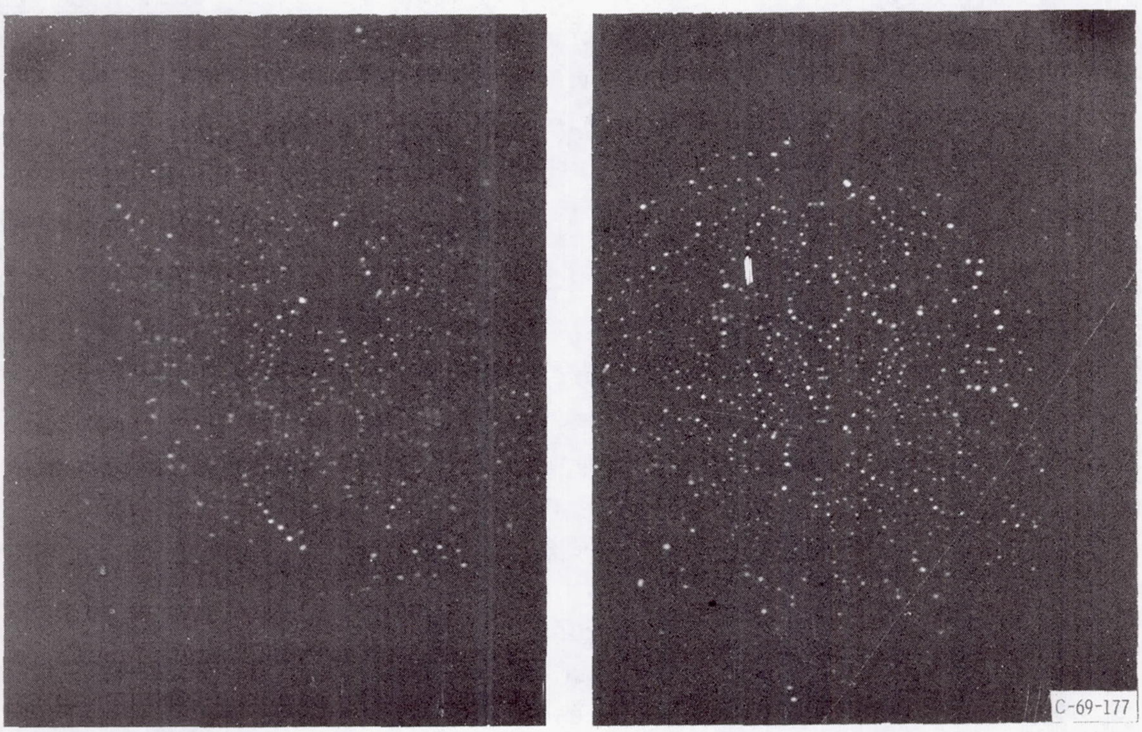
The first assumption is reasonable as can be determined from the experimental curves which appear to be quite linear in the low-temperature region.

The second assumption was made primarily for the sake of simplicity. However, the results are not very sensitive to the selection of a low-temperature expression, and the fact that the experimental slope in the low-temperature region does not vary appreciably with change of radius is not in conflict with this choice.

Using the expression,

$$T = 442\ F_o - 1.87\times10^3 \tag{3}$$

therefore, to describe the low-temperature process, and equation (1) to describe the high-temperature process, the T_{max} tip radius relation may be calculated. The constants in equation (3) were obtained by fitting it to the experimental low-temperature data for the 525-Å (525×10^{-10}-m) radius tip. Here, the field was assumed to be 4.355 volts per Å (4.355×10^{10} V/m) for an applied voltage of 15 540 volts and a tip radius of



(a) Tip temperature, 19 K.

(b) Tip temperature, 5 K.

Figure 5. - Field-ion micrograph of 270 Å (270×10^{-10} m) radius tip.

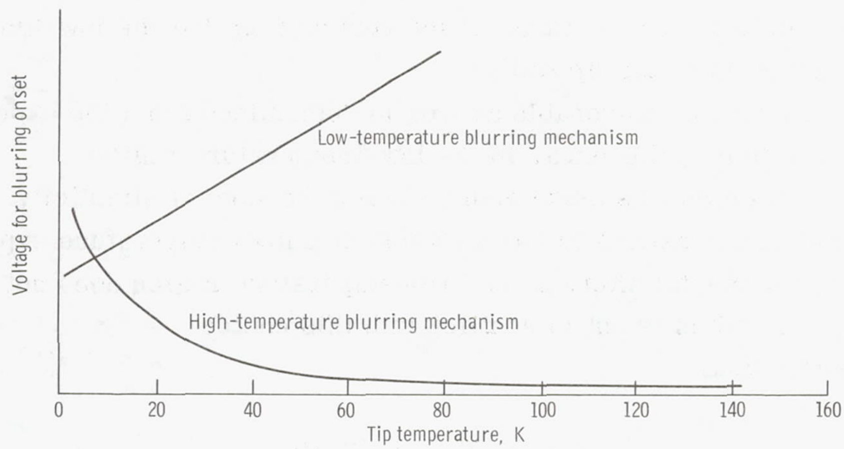


Figure 6. - Hypothetical blurring voltage versus tip temperature curve for (110) plane. Note onset of high-temperature mechanism at low temperature.

525 Å (525×10^{-10} m) (fig. 3). The results are shown in figure 4 where the agreement with experiment can be seen.

It was thought that this model might also be used to explain why the (110) plane images poorly at all but the lowest temperatures (i.e., $T \approx 5$ K). This imaging failure is illustrated in figure 5. In figure 5(a) at 19 K, note the absence of detail in the central (110) plane as compared with figure 5(b) at 5 K. Both micrographs are of the same tip. In figure 5(a), it was not possible to bring in the (110) detail by varying the applied voltage.

The thought occurred that perhaps one or the other of the blurring mechanisms might act to obscure the (110) plane at all but the lowest temperatures. Specifically, we surmised that perhaps the high-temperature blurring mechanism in the (110) plane might come into play at low temperatures, thus preventing raising the field to values that would give a well-resolved image. An example of what was pictured is shown in figure 6. Here, only at very low temperatures can the field be raised to good image quality values before the blurring mechanism takes over.

These ideas, however, were put to rest when blurring voltage versus tip temperature measurements were performed for the (111) and the (110) planes of the same tip. The results are shown in figure 7 where it can be seen that the blurring mechanisms on the (110) plane are well behaved.

From these negative results it can be concluded that the (110) plane failure mechanism is not the premature onset of a blurring mechanism and that the cause for the failure should be sought in the imaging process itself.

The behavior of the field-temperature curve in the region of 5 K requires some comment. If, as has been suggested, the low-temperature process can be described by a transit-time mechanism, then the blurring field should approach zero as the tip tempera-

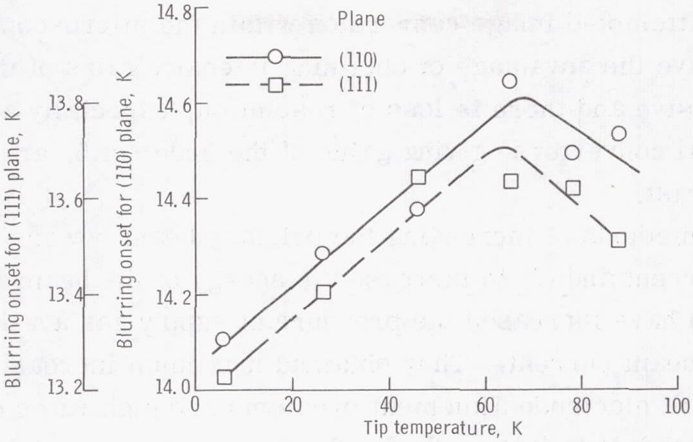


Figure 7. - Blurring voltage versus tip temperature for (110) and (111) planes and for same tip.

ture approaches zero. Because the proposed low-temperature blurring mechanism is so closely related to the imaging process, the best image field should also approach low values as the tip temperature is decreased. It would follow that, at some low temperature, the imaging field would be low enough to permit the imaging of weak materials such as copper that have not yet been satisfactorily imaged.

The experimental curves, however, in the low-temperature region, show no indication, down to 5 K, of an abrupt field dropoff with decreasing temperature. In fact, the data suggest a threshold field for blurring onset at zero absolute temperature.

To account for this, one might take into account the contribution of the large zero-point motion of helium.

Image Intensification

One of the main difficulties in field-ion microscopy is that the image intensity is so low that complete dark adaption of the eyes is necessary in order to see the image on the screen. Thus, photographic exposure times of the order of tens of minutes are necessary to record the images. Hence, there has been a continuing search for methods of increasing the intensity of the field-ion image.

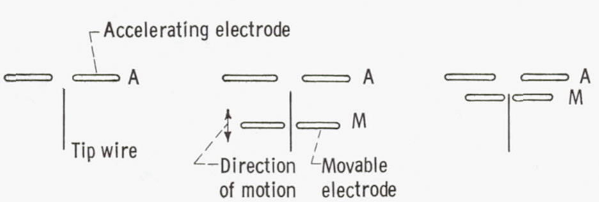
Attempts to amplify the output intensity of the field-ion microscope may be divided into two categories: (1) conversion of the primary ion beam into an electron beam, the power of which is subsequently amplified and (2) amplification of the primary ion-beam power.

In the first category are the efforts of McLane, Muller, and Nishikawa (ref. 8) and Brandon, Ranganathan, and Whitmell (ref. 9), who have used external image converters, optically coupled to the microscope, and von Ardenne (ref. 10) and Brandon et al. (ref. 9), who have attempted image conversion within the microscope envelope. The external amplifiers have the advantage of obtaining intensity gains of the order of 1000, but they are very expensive and there is loss of resolution, especially at low input light levels. The internal converters, giving gains of the order of 5, are bothered by loss of resolution and contrast.

There are two methods of increasing the primary beam power. These are (1) to increase the beam current and (2) to increase the energy of the beam ions. Waclawski and Muller (ref. 11) have increased the pressure of supply gas available to the emitter and thus increased beam current. They obtained maximum increase in intensity of the order of 50. Difficult electrode alinement problems and high rates of screen phosphor degeneration are attendant to this method. Furthermore, current amplification systems are not applicable to tip radii that are so small that their picture voltages are below the threshold for phosphor excitation.

The suggestion of increasing ion energy by postacceleration was first made by Müller (ref. 1) and carried out by Brandon et al. (ref. 9). Typical gains in exposure time of the order of 5 were reported, but, because the ion beam passed through an accelerating grid, at least 30 percent of the ion beam was lost to the grid, and the resolution was decreased.

This report describes another method of intensifying the image by increasing the energy of the beam ions (ref. 12). It is based on the relation between the applied voltage, the field at the tip, and the geometrical arrangement of the electrodes. The normal geometry in an ion microscope is as in figure 8(a) and may be described approximately as a point emitter and a planar accelerating electrode. If, however, another electrode at the same potential as the tip is introduced into the system (M in fig. 8(b)), the voltage-field relation may be changed drastically. Let us consider the limits as M is moved. If M is placed a large distance down the shank of the tip wire, the field at the cap of the tip is as it would be if M were absent. If M is moved up the shank to a position such as that in figure 8(c), the field at the cap of the tip is decreased to approximately that



(a) Normal configuration. (b) Configuration with movable electrode introduced. (c) Position of movable electrode for minimum field at tip surface.

Figure 8. - Electrode configurations.

at M. Thus, the field at the cap of the tip can be varied through a large range while the voltage on the cap remains constant. An ion produced at the emitter surface will be accelerated to the energy qV, where q is the charge on the ion and V is the applied voltage, regardless of the field at the tip. Therefore, even the smallest tip can be imaged at a brightness determined by the selection of an accelerating voltage that is limited only by the electrical breakdown characteristics between A and M, which is of the order of 100 kilovolts (ref. 13). There should be no limit to the use of extremely small tips other than the ability to make them.

Figure 9 is a photograph of a 220-Å (220×10^{-10} m) tungsten tip taken by using the variable-potential method with an applied voltage of 20 kilovolts. Normally, in a conventional microscope, this tip would have been imaged at about 4.5 kilovolts. The exposure time has been reduced by a factor of 90, while the resolution has remained unchanged.

In certain unusual cases, this phenomenon occurs without the use of an auxiliary electrode. One of these is described by Komar and Shrednik (ref. 14). As the result of an electrical discharge, the tip is ruptured, the surface of the tip being left with peaks or hills of very small radius of curvature. The remainder of the tip surface or stub acts as the electrode M and modifies the field on the peak so that intensity amplification takes place.

Another case is the phenomenon of "water etch" as described by Müller (ref. 1), where a protrusion also results. Here again the portion of the tip behind the protrusion modifies the voltage-field characteristics so that image intensification occurs. Müller (ref. 1) has reported imaging water-etched tips with radii as small as 50 Å (50×10^{-10} m).

The use of gases other than helium in the microscope has the disadvantage that the phosphor response of these gas ions is small at normal operating voltages. It should be

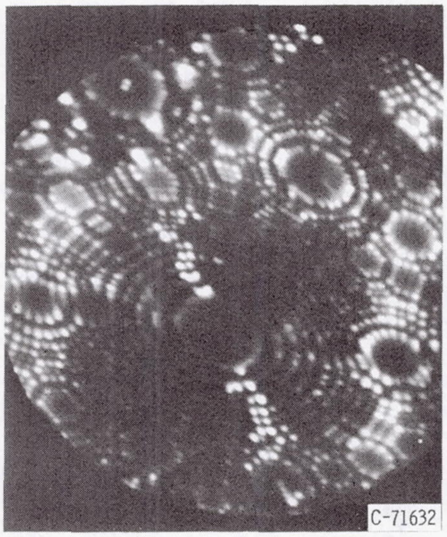


Figure 9. - Approximately 220 Å (220×10^{-10} m) tungsten tip at 20 kilovolts and 78 K.

possible with the variable-potential method of image intensification to operate the microscope with gases of lower ionization potentials than have yet been used. This would permit viewing weak-tip materials that as yet are not possible to image.

As was brought out by Brandon (ref. 9), the greater penetration of high-energy ions into phosphor spreads the phosphor damage over a larger volume, and thus the rates of phosphor degeneration in ion energy-amplification systems are not as great as in similar operation with current amplification systems. Using high voltages does, however, shorten the life of the phosphor screen, and at this laboratory a binderless settling technique such as that described by Young (ref. 15) has been used to change the screen phosphor quickly and easily.

The one difficulty encountered with the variable-potential amplification system was that the field along the shank of the tip wire was reduced to such a degree by the electrode configuration that the "water-etch" effect described by Müller (ref. 1) was very active unless very clean vacuums or low temperatures were used.

Low-Temperature Operation

It seems appropriate here to say a few words about the image quality observed at low temperatures.

(1) It has been observed that the resolution is greatest at the lowest temperatures obtained, that is, 5 K. Figure 5 gives an example of the resolution change between 19 and 5 K, respectively, for a 270-Å (270×10^{-10} m) radius tip. This is not in agreement with previously published results (ref. 1) which report 21 K as the temperature for maximum resolution.

(2) The (110) plane, which is poorly resolved if resolved at all, at higher temperatures, becomes well resolved at temperatures close to 5 K (see fig. 5).

(3) Although the intensity increases at the low temperature, the contrast seems to fall off somewhat.

(4) The differences in resolution across the tip at a given field, due to differences in local radii of curvature, seem to decrease at 5 K. Thus the entire tip can be imaged at more nearly the same applied voltage. This is no doubt related to item (2) concerning the (110) failure.

SUMMARY OF RESULTS

The results of these investigations into the resolution and image-intensity of the field-ion microscope can be summarized as follows:

1. The experimental blurring voltage versus temperature data indicate that the blurring current consists of ions that were once in thermal contact with the surface. The data seem to be satisfactorily explained by a two-mechanism theory: a transit-time mechanism at low temperatures and an orbit-time mechanism at higher temperatures.

2. The failure of the (110) plane to image at any but the lowest temperatures (~5 K) is due, not to the action of the blurring mechanisms, but to the process of image formation itself.

3. By use of the variable-potential method of increasing the intensity of the field-ion image, it is possible to reduce photographic exposure times by several orders of magnitude.

4. Maximum resolution in the field-ion microscope is reached at the lowest temperatures obtained in these experiments (~5 K).

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, January 8, 1969,
129-03-15-01-22.

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