# Electron Emission from a Lattice Step on Clean Tungsten*

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Electron emission has been observed from the edges of the outermost atom layer of a (110) crystal face of clean tungsten. A bright ring appears in the (110) area of the emission pattern on a pulsed $T$-$F$ emission electron microscope when current densities exceeding $10^7$ amp/cm$^2$ are drawn. When the emitter is maintained at an elevated temperature, $T \geq 1700°K$, the ring decreases in radius and eventually disappears. This is followed by the appearance of a new ring at the periphery of the dark (110) region, making possible the observation of a sequence of such collapsing rings. An experiment is reported which associates the ring collapse with the loss of an atom layer from the cathode tip, causing a change in its length, which is equal on the average to the interplanar spacing of the (110) crystal direction. This association proves that electron emission is observed from a lattice step and provides a convenient means for the study of the corresponding processes by which cathode material is transported.

## INTRODUCTION

THE techniques of pulsed $T$-$F$ emission microscopy[1] have been employed in the present work to observe effects of atom transport processes active on the clean tungsten cathode surface at elevated temperatures. These processes have long been known to dull and shorten field emitters.[2,3]

An emission pattern detail is shown to originate in electron emission from the edge atoms of a lattice plane of atomic thickness. Further, the transport processes which dull the emitter are shown to remove such planes from its tip in a manner which is detectable through the pattern detail.

The ability to identify and trace the progress of a lattice edge of atomic thickness may find a number of uses. For example, an accurate measurement of length change may provide means for calibrating the magnification and resolution of commercial electron microscopes. Also, the ability to resolve an atomic lattice step may prove useful for crystal surface studies where present optical interferometric methods[4] have been limited to 15 A step-height resolution. It is also clear that the methods herein discussed may be used in quantitative studies of the transport mechanisms occurring on small, heated metal objects. Subsequent studies, to be reported later, indicate that the observed process is surface migration, and provide a measure of the constants involved in that process.

Similar emission pattern effects have been observed by others[5,6]; however, direct proof of the resolution of

electron emission from the edge of an atomic plane has not been found in the literature.

This paper offers the first conclusive proof that electron emission is observed from a surface irregularity of atomic dimensions. It would appear that these techniques may be extended to determine whether emission is observed from the single atom or molecular site, about which there has been much speculation, but as yet no adequate proof.[7-9]

## DESCRIPTION OF PHENOMENA

A discussion of the difference between emission patterns obtained from tungsten during pulse and steady state operation will help to clarify the present methods. Figure 1 shows the contrast between (a) the emission pattern from clean monocrystalline tungsten at high current density $(J = 5 \times 10^7$ amp/cm$^2)$ obtained with pulsed electric fields, and (b) the pattern at low current density $(J = 25$ amp/cm$^2)$ obtained with steady state fields. In (b) the crystal planes with low Miller indices can be correlated with the large dark regions of the pattern in a well-known manner,[10] and correspond to the regions of high effective work function and hence of low electron emission. Recent measurements[6] show
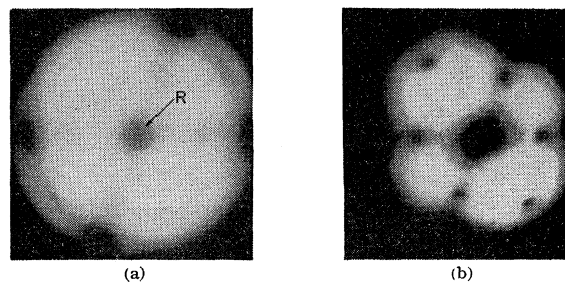


(a)                              (b)

FIG. 1. Photographs of electron projection patterns from a clean, monocrystalline tungsten emitter under the conditions of (a) high current density, $J = 5 \times 10^7$ amp/cm$^2$, during pulse $T$-$F$ operation (note the ring at $R$ in the central dark area); and (b) low current density, $J = 25$ amp/cm$^2$, typical steady-state field emission.

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[1] W. P. Dyke and J. P. Barbour, J. Appl. Phys. (to be published).

[2] E. W. Müller, Z. Physik **126**, 642 (1949).

[3] C. Herring, in *Structure and Properties of Solid Surfaces*, edited by R. Gomer and C. S. Smith (University of Chicago Press, Chicago, 1953), pp. 50, 63.

[4] Ajit Ram Verma, *Crystal Growth and Dislocations* (Academic Press, Inc., New York, 1953).

[5] E. W. Müller, 16-mm Film, Tagesztg. Deut. Physik. Gesell. September 1951, in Karlsruhe (unpublished).

[6] E. W. Müller, J. Appl. Phys. **26**, 732 (1955).

[7] E. W. Müller, Z. Naturforsch. **59**, 473 (1950).

[8] R. Gomer and D. A. Speer, J. Chem. Phys. **21**, 73 (1953).

[9] J. A. Becker, J. Chem. Phys. (to be published).

[10] E. W. Müller, Z. Physik **106**, 541 (1937).

the ratio of current density in the brightest regions of the clean tungsten pattern to that in the dark central (110) plane to be greater than 16 000/1 at the relatively low fields $(2.5 \times 10^7 < F < 3.5 \times 10^7$ v/cm) generally employed in steady state emission. It becomes apparent that observation of emission in the (110) crystal direction under such low fields is difficult and requires shielding the phosphor from scattered light and other excitation sources including x-rays, secondary electrons, and negative ions. Müller[6] reports this accomplishment in a tube where a metal anode shielded the phosphor from all electron emission except that originating in the central (110) region.

The pattern of Fig. 1(a) differs from that of 1(b) in several respects. First, the emission occurs over an increased area as evidenced by the spreading of the emission to crystal planes further from the central (110) region and by the decrease in the area of the dark "holes." Second, the current density becomes more uniform over the pattern as evidenced by the decrease in contrast between the highest and the lowest current density regions.[11] This can be attributed to two causes: (1) a decrease in the ratio of current densities between the low and high work function regions at high fields, as compared with the ratio at low fields as predicted by the Fowler-Nordheim field emission theory,[12] and (2) electron space-charge effects[13] which tend to prevent further current density increase in "bright" regions while permitting such increase where current density is still relatively low. These two effects decrease the pattern contrast and make it possible to observe detail that may occur in the low-index planes such as at $R$ of Fig. 1(a), where a bright ring[14] of emission is apparent near the periphery of the central (110) dark region. When patterns such as Fig. 1(a) are observed by the methods of pulsed $T$-$F$ emission microscopy as described in reference 1, and when a cathode temperature in the range 1700° to 3000°K is used for tungsten, the ring $R$ in the figure is seen to decrease in diameter and eventually to disappear; later another ring will appear and the process of ring collapse can be repeated. Such a sequence is illustrated in Fig. 2. The ring initially becomes apparent at the periphery of the dark (110) area as illustrated at $R$ in Fig. 2(a), and as the emitter temperature is kept constant the ring continuously decreases in diameter as shown in Figs. 2(b) and (c) until it finally vanishes as at (d). Following the disappearance of the ring, the 110 region remains devoid of detail for a short period of time, after which another ring can be resolved at the periphery, to be followed by
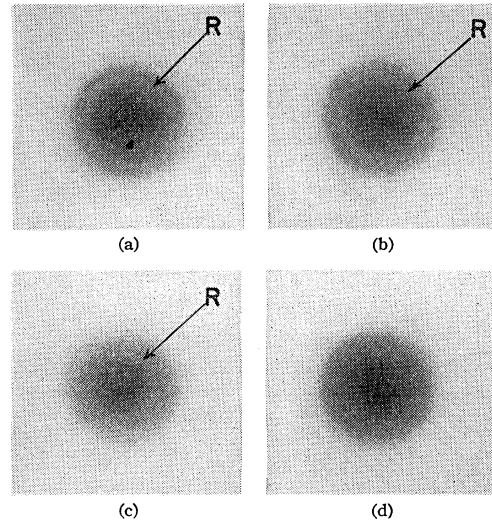


FIG. 2. Photographs of the central 110 plane on the emission pattern showing successive stages of ring collapse [(a) through (d)] corresponding to the dissolution of the outermost atom layer from the 110 crystal plane of the heated tungsten tip, viewed by pulsed $T$-$F$ emission electron projection microscopy.

the shrinking sequence described in the foregoing. If a constant, high emitter-tip temperature is maintained, the sequence will recur at nearly uniformly spaced intervals of time. The repetitive pulsing technique is used only for purposes of viewing the pattern; at the pulse lengths and repetition rates used herein, i.e., 1 μsec and 30 to 1000 pps (pulses per second) respectively, the average value of the electric field is negligible.

The number of such collapsing rings per unit of time, herein referred to as "ring rate," increases rapidly with increasing tip temperature, decreases with increasing radius of curvature of the tip, and decreases with increasing polar angle for a given type of crystal plane. It is also strongly dependent on the state of cleanliness of the tip surface; for example, less than a monolayer of carbon will impede the rate of ring collapse on tungsten. A ring may be "frozen-in" or held at any diameter by reducing the tip temperature below 1000°K.

## PROPOSED ORIGIN OF EMISSION IN THE RING AND THE MECHANISM OF RING DISSOLUTION

The existence of the ring can be explained on the basis that the corresponding electron emission originates at the edge atoms of the top atom layer of the (110) face as illustrated in Fig. 3. Geometrical considerations show the electric field at the edge atoms to be greater than that existing at the smooth, tightly packed surface of the plane; these edge atoms may also present a lower effective work function due to increased surface roughness.[15] Both effects result in increased field emission as compared with that of their surface neighbors. Thus a ring of emission would be expected to

[11] Dyke, Trolan, Dolan, and Grundhauser, J. Appl. Phys. 25, 106 (1954).
[12] R. H. Fowler and L. W. Nordheim, Proc. Roy. Soc. (London) A119, 173 (1928).
[13] Barbour, Dolan, Trolan, Martin, and Dyke, Phys. Rev. 92, 45 (1953).
[14] Not to be confused with the "ring and tilt" associated with vacuum arc initiation. [Dyke, Trolan, Martin, and Barbour, Phys. Rev. 91, 1043 (1953).]
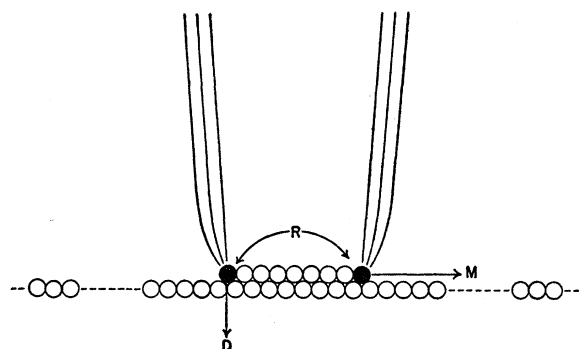
[15] R. Smoluchowski, Phys. Rev. 60, 661 (1941).

FIG. 3. Cross section of idealized emitter tip showing two outermost atom layers. Emission forming pattern ring originates in the vicinity of edge atoms ($R$) of top layer. Edge atoms at $R$ are least tightly bound and when thermally agitated may be removed by surface migration as at $M$ or by volume diffusion into vacancies in the metal lattice as at $D$.

occur and also to be resolved in the emission pattern detail, provided the diameter of the top lattice step was sufficiently smaller than that of the underlying step to be resolved by the field emission microscope. The resolving power of the device is calculated to be about 40 A for the smooth cathode[16] and 3 A in the presence of surface roughness.[17] If an emitter tip having a radius of about 1000 atoms, a size appropriate for this study, is idealized as a hemisphere, several single lattice steps should be individually resolvable with the field emission microscope; however, the shape assumed by the cathode at elevated temperatures is such that the low-index planes are extended in size and lattice steps become bunched in the region between the simple planes. A resolvable separation between the edge of the top atom layer and underlying layers occurs occasionally following flash heating of the tip, or periodically on a continuously heated emitter.

If the emitter tip is maintained at an elevated temperature, the thermal energy of the atoms may become sufficient to break some of the bonds of the less tightly bound edge atoms, as at $R$ of Fig. 3. These atoms may then proceed across the plane by surface migration, as represented by the arrow at $M$, or they may, by the process of volume diffusion, follow a path $D$ into the interior of the metal. The rates of these transport processes have been discussed by Herring.[3] At high temperatures evaporation can also remove atoms; however, at the temperatures employed in this work, the latter process can be neglected. The removal of atoms from the lattice step results in a decrease in the diameter of both the top atom layer and its associated emission ring.

The proposed explanation of the ring formation and removal assumes that electron emission from the edge of a single atom layer is resolved. The following experiment establishes that assumption and gives assurance

[16] E. W. Müller, Z. Physik 120, 270 (1943).
[17] D. J. Rose, Phys. Rev. 98, 1169 (1955).

that rings, under the conditions of the present experiment, do not correspond to multiple atom layers.

## EXPERIMENTAL PROCEDURE AND RESULTS

The purpose of the experiment was to observe a sufficient number of successive collapsing rings on the emission pattern from a single tip that, if one assumes each ring to be associated with the removal of one or more atom layers, the resulting change in emitter tip length would be large enough to be accurately measured by other means. A convenient method of such measurement is by use of electron micrographs where a resolution of 100 A requires a total of about 400 rings for a reasonably accurate measurement of emitter length change.

In order to have a reference point from which to measure the emitter-tip length, it is necessary to have some identifying mark or characteristic in the vicinity of the tip, which will remain fixed relative to the bulk of the emitter during heat treatment. Desirable reference markings were produced when emitters were fabricated from tungsten wire composed of small crystals about $10^{-3}$ to $10^{-4}$ cm in length. Emitters etched from this stock were smooth and uniform following the etching process. They were then heated in good vacuum at a temperature of 2000°C from two to ten minutes for the dual purposes of forming indentations at crystal boundaries as at point $P$ of the tip shadowgraph of Fig. 4(a), and to smooth and round the tip to the extent indicated in the same figure. Further heating to clean and outgas during the final evacuation process was held to a minimum to reduce
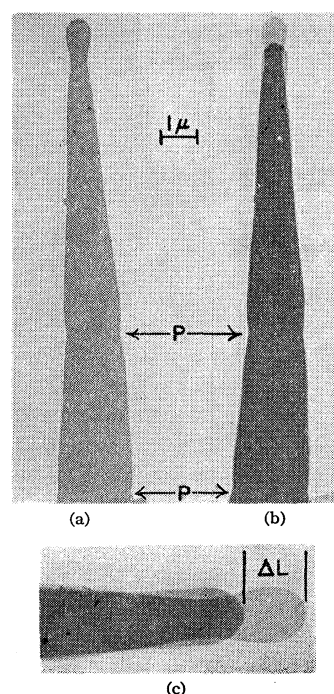


(a)          (b)

(c)

FIG. 4. Electron microscope shadowgraphs of the profile of emitter X160. $A$, before operation, with length-reference point indicated at $P$. $B$, superposition of shadowgraphs obtained before and after operation with matched reference indentations at $P$. $C$ shows an expanded view of the superimposed tips; $\Delta L$ is the decrease in emitter length resulting from the collapse of 3000 rings.

undesired changes in the emitter length. The double indentations in the shank of the emitter shown in Fig. 4(a) serve as double reference points and permit detection of possible reference point shift through grain growth. No change in grain size or movement of grain boundaries was noted during the experiment. In other cases, curved emitter blanks afforded an independent reference for the measurement of length change.

After fabrication, electron micrographs were obtained and each emitter was mounted in an electron projection tube,[11] evacuated as previously described[18] and sealed off at a pressure of about $10^{-8}$ mm of Hg. After seal-off and prior to operation, tantalum gettering decreased the static, chemically active gas pressure to about $10^{-12}$ mm of Hg.

During $T$-$F$ emission, the total number of collapsing rings was determined either by direct count, as was done in the case of emitter No. X127 of Table I, or by observing the rate at known intervals of time and adding the individual rate-time products. After the collapse of some 2000 to 3000 rings had been observed, the emitters were removed from their envelopes and again micrographed at a known magnification. The shadowgraphs of each emitter obtained before and after the ring count were then adjusted to the same magnification and carefully superimposed at the reference notches as at $P$ in Fig. 4(b) and the change in emitter length measured. Table I shows a column of values of the measured change in length for three emitters with an uncertainty estimated at $\pm 10\%$ which includes errors due to microscope calibration and matching the micrographs as the principal sources. Table I also shows for comparison purposes the predicted change in length for each emitter, assuming that each collapsing ring is associated with the removal of a single atom layer from the (110) plane of the tip. The interplanar spacing in the (110) crystal direction for tungsten is $2.23 \times 10^{-8}$ cm; thus, for example, the removal of 2500 atom layers from the tip amounts to a length change of $5.6 \times 10^{-5}$ cm. Good agreement is noted between the measured and predicted columns of the table.

## CONCLUSION

The foregoing describes conditions under which the emission from the edge atoms of an atomic plane on the (110) face of clean tungsten is resolved in the field emission microscope. The results show that the collapse of an emission ring of the type described is associated

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[18] W. P. Dyke and J. K. Trolan, Phys. Rev. **89**, 799 (1953).

TABLE I. Number of collapsing rings and change in emitter length, for three emitters.

| Emitter designation | Number of rings | Change in emitter length — Predicted value on the basis: 1 ring to 1 atom layer | Change in emitter length — Measured value from emitter shadowgraphs |
|---|---|---|---|
| X160 | 3000 | $6.7 \times 10^{-5}$ cm | $(6.4 \pm 0.6) \times 10^{-5}$ cm |
| X127 | 1960 | $4.4 \times 10^{-5}$ cm | $(5.0 \pm 0.5) \times 10^{-5}$ cm |
| Q218 | 2500 | $5.6 \times 10^{-5}$ cm | $(5.7 \pm 0.6) \times 10^{-5}$ cm |

with the dissolution of a single layer of atoms from the plane in question.

Since, by these means, one can identify the removal of such minute quantities of material as a single layer or a portion of a single layer from an emitter tip, one has a convenient tool for studying the mechanisms of atom transport and their dependence on various parameters. The advantages of this technique are several: (1) transport measurements can be made on an emitter while its gross geometry, which through mean surface curvature mainly determines the transport rate,[3] is essentially constant; (2) the transport process is made continuously visible through electron emission resulting from a high electric field, yet undesired effects due to the field can be avoided by use of pulsed operation at a very low duty-cycle[1]; (3) the temperature necessary for the transport of detectable amounts of tungsten cathode material also causes evaporation of most surface contaminants, so that the surface is maintained in a relatively clean condition.

Direct observation of a virtually instantaneous transport rate may be useful in several related problems. By measuring the dependence of the ring rate on temperature one may deduce the activation energy of the process involved; such a result should be readily accessible for several metals. Rates of transport may be expected to depend not only on temperature but on temperature gradient, surface curvature and its gradient, applied field, state of cleanliness or contamination, and perhaps other factors. Conversely, study of the rate during controlled variation of these parameters may clarify some of the present obscurity concerning the mutual relationship of the corresponding mechanisms.
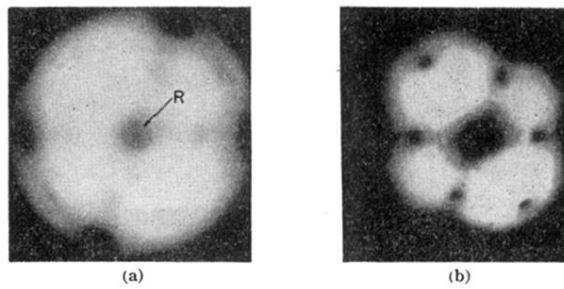
## ACKNOWLEDGMENTS

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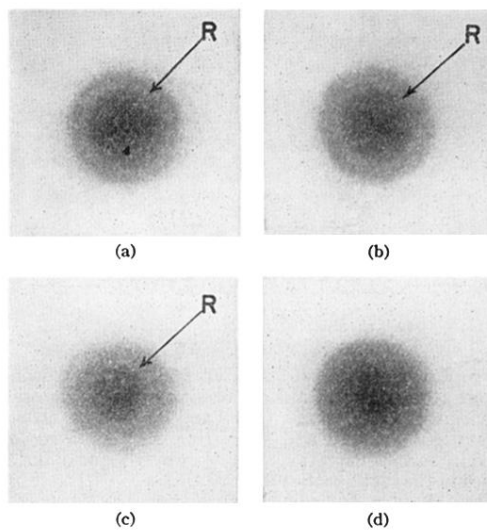
FIG. 2. Photographs of the central 110 plane on the emission pattern showing successive stages of ring collapse [(a) through (d)] corresponding to the dissolution of the outermost atom layer from the 110 crystal plane of the heated tungsten tip, viewed by pulsed *T-F* emission electron projection microscopy.
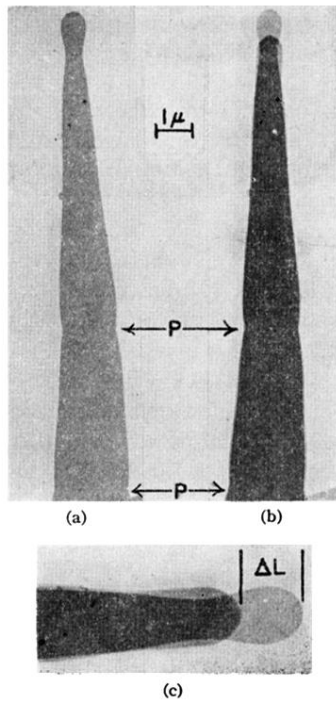
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