

The Photoemission of Tungsten in the Region of Predicted Schottky Deviations*

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Experimental evidence for the existence of deviations from the photoelectric Schottky line theoretically predicted by Guth-Mullin has been obtained. In the phase of the periodic deviations an averaging of data yields for example a maximum at 219 ± 18 (v/cm) $^{1/2}$ and a minimum at 278 ± 16 , in agreement with theoretical values of 215 and 285, respectively. The amplitudes of the deviations are 0.3–1.5 the predicted values with smaller than theoretical values predominating.

A noted difference between data from ac and dc heated filamentary cathodes is considered in relation to the effect of emission surface roughness on electric field strength calculations. Also, patch theory has been applied to the photoelectric data, supplying evidence of patches of about 6 microns in extent for dc heated wires. An electron microscope study of the tungsten filament emission surfaces supports the conclusions.

The influence of more recent theory on the interpretation of the experimental results is briefly discussed in an appendix.

I. INTRODUCTION

GUTH and Mullin,¹ employing an electric-field-dependent electron transmission coefficient, theoretically predicted a photoelectric analog of the well established^{2–6} deviations of thermionic electron emission from the straight Schottky line. For photoemission, their field dependent photocurrent i is, to a good approximation,

$$i \propto a + bE^{3/2} + cE - \frac{(d + fE^{3/2}) \cos u}{(g + jE^{-3/2} + pE^{-1/2})^{1/2}}, \quad (1)$$

where

$$a = (\pi kT)^2 / 6 + h^2(\nu - \nu_0)^2 / 2,$$

$$b = h(\nu - \nu_0)e^{3/2},$$

$$c = e^3 / 2,$$

$$d = W_a^{1/2} h(\nu - \nu_0) / 2,$$

$$f = W_a^{1/2} e^{3/2} / 2,$$

$$g = (kT)^{-2}$$

$$j = (300e)^{3/2} [\pi^2 + (\gamma + 2 \ln 2)^2] / 16q^3,$$

$$p = \pi(300e)^{3/2} / 2kTq^{3/2},$$

$$q = h^2 / me^2,$$

$$u = (4/3q^{1/2})(300e)^{1/2} E^{-1/2} - 2W_a^{-1/2}$$

$$+ \tan^{-1} \left[\frac{W_a^{1/2}}{4} \right] - \tan^{-1} \left[\frac{(\gamma + 2 \ln 2)(300e/q^2)^{3/2} E^{-3/2}}{\pi(300e/q^2)^{3/2} E^{-3/2} + (kT)^{-1}} \right],$$

e = electronic charge,

E = applied electrostatic field,

γ = Euler's constant (0.5772),

ν = incident frequency,

ν_0 = threshold frequency, and

* Assisted by the U. S. Office of Naval Research.

¹ E. Guth and C. J. Mullin, Phys. Rev. **59**, 575 (1941); **59**, 867 (1941); **61**, 339 (1942); **60**, 535 (1941).

² R. L. Seifert and T. E. Phipps, Phys. Rev. **56**, 652 (1939).

³ D. Turnbull and T. E. Phipps, Phys. Rev. **56**, 663 (1939).

⁴ W. B. Nottingham, Phys. Rev. **57**, 935 (1940). See also the last reference of footnote 1.

⁵ Munick, LaBerge, and Coomes, Phys. Rev. **80**, 887 (1950).

⁶ E. A. Coomes, Phys. Rev. **85**, 392 (1952).

W_a is the barrier height, which for the assumption of one free electron per metal atom is 10.33 ev for tungsten.

For a field-independent transmission coefficient (normal Schottky effect) the theory shows that the last term of Eq. (1) is zero, yielding

$$i_0 \propto a + bE^{3/2} + cE. \quad (2)$$

The experimental investigation of Eq. (1) for tungsten is the subject of the present paper, which appears to be the first experimental study of this effect for a metallic element. Carroll and Coomes⁷ have done similar work on BaO, finding qualitative agreement with the Guth-Mullin theory.

To interpret the experimental results it is convenient to plot $\Delta i / i_0$, where Δi is the last term of Eq. (1) and i_0 is given by Eq. (2). The reference photocurrent i_0 as a function of $E^{3/2}$ is shown by Fig. 1 for two important incident light frequencies. For the present experimental conditions the parabola of Eq. (2) is practically a straight line for the fields plotted.

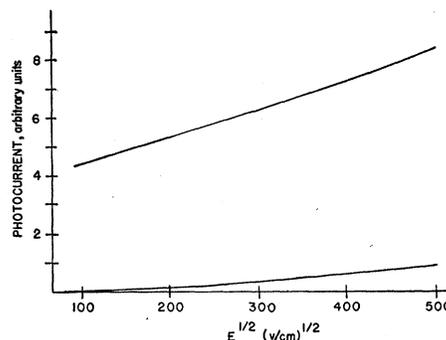


FIG. 1. Photoemission vs the half-power of the applied electric field according to Eq. (2). The upper plot is for the present experimental conditions ($T = 293^\circ\text{K}$ and $h(\nu - \nu_0) = 0.368$ ev), with the proportionality constant of the theory taken as 10^4 . The lower curve is for $\nu = \nu_0$. The transmission coefficient of the barrier is considered constant.

⁷ P. E. Carroll and E. A. Coomes, Phys. Rev. **85**, 389 (1952).

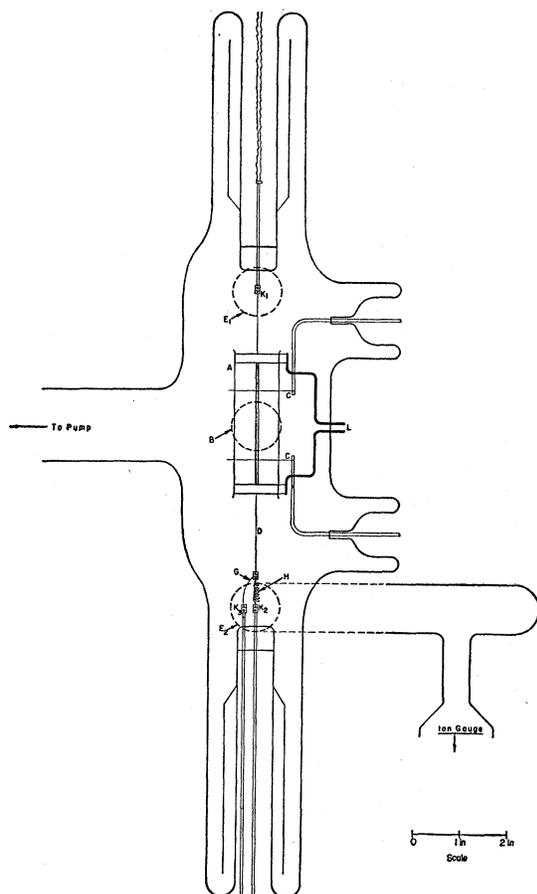


FIG. 2. Phototube. *A*, 3-in. tantalum cylindrical anode of internal diameter 0.75 in. with 1-mm slit in wall; *B*, quartz window in wall of main glass tubing; *C*, 0.015-in. tungsten filaments (for outgassing *A* by electron bombardment) welded to two-wire presses; *D*, 0.004-in. tungsten filamentary cathode; *E*₁ and *E*₂, access tubes for changing filament *D*, with *E*₂ connected to ionization gauge; *G*, 0.020-in. nickel wire for by-passing spring *H* during cathode (filament) outgassing; *H*, 0.015-in. coiled tungsten spring for filament tautness; *K*₁, *K*₂, *K*₃, nickel chucks for fastening wires to press leads.

The Guth-Mullin theory involves several assumptions: (1) The Fowler theory of the photoeffect, the basis of the work of Guth and Mullin, relies on the assumption that the incident frequency is near the threshold frequency. The frequency used in the present work (λ , 2537Å) differs from the threshold frequency by only 8 percent, well within the range allowed by Fowler.⁸ (2) The problem is considered as one-dimensional. This is a considerable assumption in view of the periodic structure of the metal surface. However, Herring and Nichols⁹ indicate that the assumption is at least reasonable. (3) The zero-field threshold energy difference $h(\nu - \nu_0)$ is replaced simply by the field dependent threshold energy difference $h(\nu - \nu_0) + e^{\frac{3}{2}}E^{\frac{1}{2}}$.

⁸ R. H. Fowler, *Phys. Rev.* **38**, 45 (1931).

⁹ C. Herring and M. H. Nichols, *Revs. Modern Phys.* **21**, 185 (1949).

This change, corresponding to the Schottky lowering of the potential barrier at the metal surface, is amply justified by the experimental confirmation of the thermionic Schottky effect and by the photoelectric study of Lawrence and Linford.¹⁰ (4) The surface is considered to have a constant work function.¹¹

II. EXPERIMENTAL

The evacuation and outgassing procedure used on the several phototubes was standard. The earlier tubes were sealed off containing Kemet Laboratories KIC barium core getters. However, on a suggestion of W. B. Nottingham the later tubes were sealed off without getters to avoid introducing foreign substances into the tube. After seal-off further lowering of the pressure was obtained by continuous operation of the ionization gauge.¹²

Pressures were measured with either VG-1A or Bayard-Alpert¹³ ionization gauges.

The phototube design is shown by Fig. 2. The quartz window opposite the slit in the anode enables the incident radiation (from a narrow slit) to enter the tube without appreciable scattering. Although the light is incident predominantly on one portion of the central wire filament, emission also occurs from the other areas of the cathode surface due to reflections of the radiation from the inner surface of the anode. The electric field at the surface of the central filament is calculated from the assumed ideal cylindrical geometry of the tube. For the 0.004-in. wire used, E is 37.7 V v/cm, where V is the magnitude of the applied voltage. It can be shown that the centering of the filament is not critical for the dimensions actually used.

The light source was a Hanovia SC-2537 low pressure mercury arc which is practically a monochromatic ultraviolet source of 2537Å radiation.¹⁴

For most of the work the phototube potential was provided by a bank of 45-v *B* batteries which with a specially constructed voltage divider provided a continuously variable voltage from 0 to 4500 v. The batteries gave a very steady voltage when new and well cared for but developed leakage paths to ground with the passage of time. To overcome the inconvenience of the batteries, an electronic power supply was constructed. It was a simple half-wave rectifier with a 2- μ f output capacitor and a 20-megohm filter resistor.

The photocurrent amplifier was a DuBridge-Brown dc amplifier using an *FP-54* electrometer tetrode.¹⁵ It was stable (zero drift of about 0.3 percent of the usual photocurrent per hour) and, over the range of input voltages used (0–0.25 v), quite linear. The output

¹⁰ E. O. Lawrence and L. B. Linford, *Phys. Rev.* **36**, 482 (1930).

¹¹ Deviations were studied only for regions where patch effects could be neglected.

¹² W. v. Meyern, *Z. Physik* **84**, 531 (1933).

¹³ R. T. Bayard and D. Alpert, *Rev. Sci. Instr.* **21**, 571 (1950).

¹⁴ A. H. Weber, *Phys. Rev.* **53**, 895 (1938). See also manufacturer's data.

¹⁵ L. A. DuBridge and H. Brown, *Rev. Sci. Instr.* **4**, 283 (1933).

current of the amplifier was measured either by a Leeds and Northrup wall galvanometer of sensitivity 4×10^{-9} amp/mm or by a Minneapolis-Honeywell Brown recording potentiometer. Leakage currents, mechanical vibrations, and variations of arc intensity were minimized. Arc current fluctuations were not more than 0.25 percent, with resultant photocurrent changes of less than 0.13 percent.

When the galvanometer was used for readings the method of observation was to wait for about 10 minutes after changing the voltage to obtain a practically constant background of low magnitude and then to take 7 readings of photocurrent in succession. The average of these is the value plotted on the curves. When the recording potentiometer was used it was found possible to decrease the waiting period since accurate readings could be taken from the record obtained.

III. RESULTS

Graphical analysis is the most convenient method of examining the experimental data. Figure 3 presents the essential results of the Guth-Mullin theory. The principal difficulty of working the experimental data into the form of Fig. 3 was in determining the correct Schottky line representing the theory for constant transmission coefficient, analytically expressed by Eq. (2). Since the actual experimental currents represented the sum of undeviated currents and corresponding deviations, the observed currents could not yield a unique undeviated line.

The method of analysis was to represent the data by a straight line, visually adjusted to give a good fit over the significant portion of the curve. This could be done more accurately for those curves of larger abscissa ranges than for those of limited extent. In any case the line was somewhat arbitrary. Since Eq. (2) involves an undetermined proportionality constant, the slope of the experimental line cannot be compared to the theory. After determination of the line for the undeviated current i_0 the difference Δi between the measured and the undeviated currents for a given field value was ob-

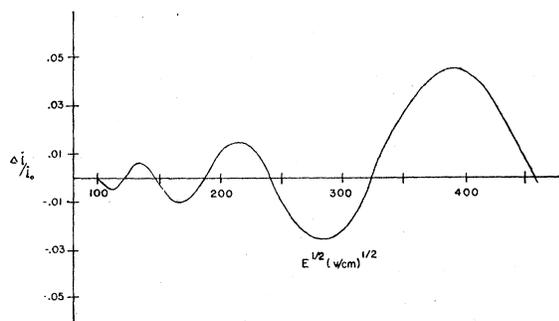


FIG. 3. Deviations, calculated by the Guth-Mullin theory Eqs. (1) and (2), from the photoelectric Schottky effect. The ordinates are the fractional magnitudes of the deviations from the constant transmission coefficient theory. The calculations have been made for 293°K and an incident wavelength of 2537Å.

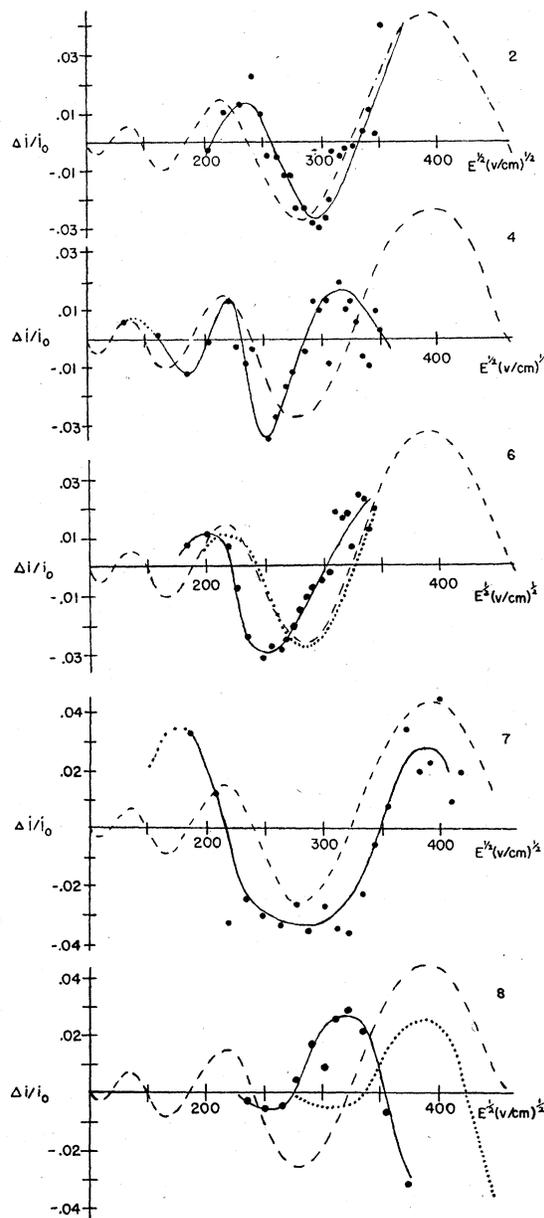


FIG. 4. Experimental results for dc-heated wires and unpolished tungsten. The dashed lines are the theoretical predictions; the dotted lines in Curves 4 and 7 are extensions of the experimental curves in ambiguous regions; the dotted lines of Curves 6 and 8 are the experimental curves obtained when the surface field is increased 1.2 and 1.5 times, respectively, to allow for surface roughness. The curves are numbered in the order in which the data were obtained, with the first three sets of data obtained from one tube and the other two from another tube.

tained. The experimental $\Delta i/i_0$ can then be compared with the theoretical curve of Fig. 3.

The essential results of the present investigation are shown in Figs. 4 and 5. The curves of Fig. 4 are for filaments that were not polished and were outgassed with dc. Curve 2 was obtained from a tube at a pressure

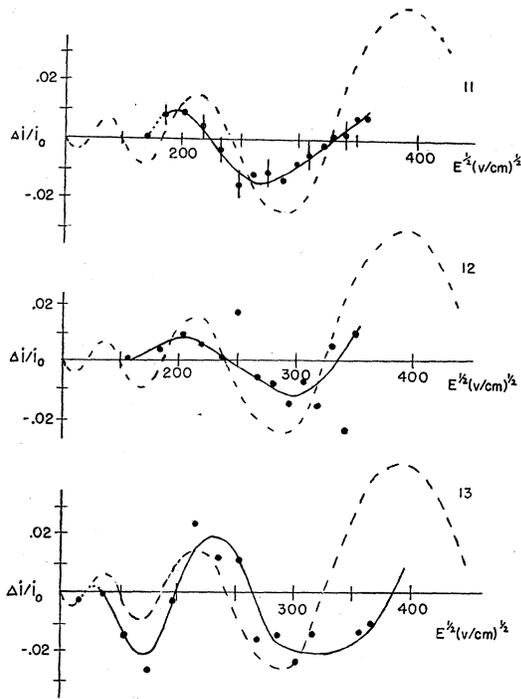


FIG. 5. Experimental results for ac-heated wires. The dashed lines are the theory; the dotted extensions of Curves 11 and 13 are for ambiguous regions. Standard deviations of the current readings are given for some of the points of Curve 11. Curve 13 is the only curve for the electronic power supply and recording potentiometer.

of 10^{-7} mm Hg or less; 4, 6, 7, and 8 were obtained from tubes at about 5×10^{-7} mm Hg.

Figure 5 shows three curves for wires manually polished and outgassed with ac. Curve 11 was obtained from a phototube at a pressure of 3×10^{-7} mm Hg, while the other two curves were taken at about 2×10^{-8} mm Hg. To indicate the accuracy of the deviation amplitudes, the fractional magnitudes of the standard deviations of important points are shown on Curve 11.

TABLE I. Observed and predicted deviations.^a

Curve ^b	Observed deviations									Agreement with theory ^c			
	0	Max	0	Min	0	Max	0	Min	0		Max		
Accurate field measurements (ac heating)													
9 ns						200	260	325			F		
10 ns						200	260	305			F		
11				170	195	225	270	330			G		
12				160	205	245	295	335			G		
13	120	127	135	170	200	230	265	315	385		G		
Inaccurate field measurements (dc heating)													
2						205	235	255	295	335	G		
3 ns						255	270	280	315	345	F		
4				160	185	205	220	230	250	285	315	350	P
5 ns						200	240	280	300	320		P	
6						170	200	225	250	300		G	
7							175	215	280	350	390	445	F
8								225	250	275	320	355	F
Theory	120	135	150	165	185	215	240	285	325	390	460		

^a All measurements are in $(v/cm)^{1/2}$.

^b ns, not shown in Figs. 4 and 5.

^c G, good; F, fair; P, poor.

It is seen that the amplitudes of the Schottky deviation are greater than the usual standard deviations of the individual points and this is generally true of all the curves obtained.

Table I gives a summary of the experimental data and the corresponding theoretical positions of maxima, minima and intercepts. The data for four runs not plotted in Figs. 4 and 5 are included. These four runs show less favorable agreement in general between theory and experiment than is indicated by the eight runs of Figs. 4 and 5. Table II contains the results of averaging the values of Table I.

The amplitudes of the deviations are not uniquely established but appear usually to be somewhat smaller than theory predicts, varying from 0.3 to 1.5 the corresponding calculated values.

IV. DISCUSSION

A. Surface Roughness

A factor of importance is the roughness of the emitting surface. In addition to changing the effective area for

TABLE II. Phase relationships of observed to predicted deviations.^a

	Experiment Average of Table I values	Theory	
		Guth- Mullin	Juenker <i>et al.</i>
First zero	120 ^b	120	108
First maximum	133±4 ^b	135	120
Second zero	148±8 ^b	150	130
First minimum	178±5 ^b	165	145
Third zero	196±19	185	160
Second maximum	219±18	215	180
Fourth zero	237±18	240	204
Second minimum	278±16	285	235
Fifth zero	324±19	325	266
Third maximum	342±23 ^b	390	315
Sixth zero	383±29 ^b	460	360

^a All data in $(v/cm)^{1/2}$.

^b Not reliable due to insufficient data.

photoemission and gas adsorption, roughness affects the magnitude of the applied electric field. Since the field strength is inversely proportional to the radius of the filamentary cathode any sharp irregularity will increase the local field.

The first sets of data (Fig. 4) obtained in this study were for filaments that were rough due both to lack of polishing and to the etching effect of dc heating.¹⁶ Later filaments (Fig. 5) were polished and heated with ac to avoid the roughness. An electron microscope study of the filaments used in this research (Callite No. 200-H) has shown that unpolished, dc heated wires are rougher than polished wires comparably heated with ac. The notable differences between the curves of Fig. 4 and those of Fig. 5 are probably due to the smoother filament surfaces of the latter. The dc roughness is ap-

¹⁶ See reference 9, pp. 202, 261.

parently time dependent for runs taken before and after heating are not consistent in the phase locations of the deviations. The electron microscope study also indicated that even moderate dc heating can change filament surfaces. Hence if the filament is dc heated between runs, the values of the applied fields for given voltages will generally be different due to the changes in the surface structure.

As an example of how this dc roughness may affect the experimental data, it is interesting to examine Curve 6 of Fig. 4. If a multiplying factor of 1.2 for the field is assumed as a correction for the roughness of the emitter, the experimental curve becomes that shown by the *dotted* line. The improved agreement with theory over a complete cycle may mean that surface roughness for dc heated filaments is a source of experimental error.

The evidence indicates that ac heated filaments are not rough enough to have surface fields materially larger than calculated.

B. Patch Effects

The tungsten filament wire used is polycrystalline. Before heating in a vacuum, its crystals are of the order of half a wire diameter or less in length, as determined by inspection under polarized light. Heating recrystallizes the wire, for examination after the heat treatment showed crystals in a wire diameter or more in length.

The crystal structure of the wire leads to the problem of patch effects.⁹ Since the various crystal planes of tungsten differ in their work functions, the presence of different planes at the surface of the metal produces patches of different work functions.¹⁷ Furthermore, the roughness of the surface affects the size of the patches exposed at any place.

Application of the theory of Herring and Nichols¹⁸ yields the result that the empirical slope of the straight line approximation for the normal Schottky effect [Eq. (2) with the cE term dropped] should be smaller than the calculated slope in the weak field region and larger in the strong field region.

Of interest in the present investigation is the location of the breaks in slope in the region between intermediate and large fields. Many of the photocurrent *versus* (applied field)³ curves obtained (from which the deviation curves of Figs. 4 and 5 are derived) have a sharp change in slope near $180 \text{ (v/cm)}^{\frac{1}{2}}$. If such a break occurs where the field satisfies the strong field criterion δ to the extent that $2E = 100(\delta\phi/\Delta y)$, and if $\delta\phi$ be taken as 0.4 v, the indicated patch diameter would be of the order of six microns.¹⁹ This agrees reasonably with the patch size indications given by the electron microscope study of the filament surfaces.

¹⁷ M. H. Nicols, Phys. Rev. **78**, 158 (1950).

¹⁸ See reference 9, pp. 204–205, 214.

¹⁹ Writing the inequality $E \gg \delta\phi/\Delta y$ (reference 9, p. 204, II.5.4) as the equality $2E = 100(\delta\phi/\Delta y)$.

V. CONCLUSIONS

The present investigation has produced good evidence for the existence of the quasi-periodic deviations of the photoemission of tungsten from the Schottky effect for a constant transmission coefficient. Errors were carefully considered and the usual causes of current fluctuations were systematically eliminated. The main dependence for the conclusions is on the statistical nature of the data. Of especial importance is the fact that the standard deviations of the current readings for the individual points near practically all of the maxima and minima are appreciably less than the amplitudes of the deviations.

The amplitude of the measured deviations is 0.3–1.5 the theoretical value, being generally smaller than theory predicts, while the phase locations averaged from the curves of Figs. 4 and 5 are in closer agreement with the theory. Thus, a maximum (average) at $219 \pm 18 \text{ (v/cm)}^{\frac{1}{2}}$ and a minimum (average) at 278 ± 16 correspond to calculated values of 215 and $285 \text{ (v/cm)}^{\frac{1}{2}}$ and intercepts at 196 ± 19 , 237 ± 18 and $324 \pm 19 \text{ (v/cm)}^{\frac{1}{2}}$ correspond to calculated values of 185, 240 and $325 \text{ (v/cm)}^{\frac{1}{2}}$.

It is particularly noted that: (1) dc heating of filaments may cause sufficient surface roughness to make the actual applied field noticeably larger than ideally calculated and to cause patch breaks in the region of fields of interest; (2) since all readings were taken for filaments with one or more layers of adsorbed gas the presence of such layers does not completely inhibit the Guth-Mullin deviation effect,⁵ though it may be a contributing cause to the small amplitudes of a number of deviation curves.

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VI. APPENDIX

A subsequent consideration of the experimental results presented above in light of the recent theoretical modifications of Juenker *et al.*²⁰ of the original theory¹ of Schottky deviations yields these conclusions. (1) The present photoelectric experiments agree somewhat better in average with the original Guth-Mullin¹ theory in the phase of the deviations, although several individual curves (such as Fig. 4, curve 8 and Fig. 5, curves 11 and 12) suggest deviation phases rather close to those of the Juenker theory which are shifted about a quarter-period relative to the older theory. Hence there remains the possibility that the proposed dc roughness factor suggested (Fig. 4, dotted curve 6 and 8) may be invalid with the correct explanation of the out-of-phase character of the experimental and

²⁰ Juenker, Colladay, and Coomes, Phys. Rev. **89**, 894 (1953); **90**, 772 (1953); and private communication.

theoretical results lying in the incorrectness as to phase locations of the old theory. (2) The present photoelectric experiments agree approximately in amplitude of the deviations with the Guth-Mullin theory with, as has been pointed out, the experimental amplitudes coming out somewhat smaller usually. The experimental amplitudes therefore are smaller than predicted by the old

theory and larger than by the Juenker theory. (3) The present photoelectric experiments agree quite closely with the Juenker theory in the magnitude of the difference between positions of successive zero deviation points; 0.9π comes out of the experiments as the average value of the half-period of the appropriate parameter as against π in the theory.

The Storage of Energy in Silver Activated Potassium Chloride*

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Crystals of KCl-Ag and NaCl-Ag have been excited by x-ray irradiation. The photostimulated light yield of the ultraviolet emission band has been observed simultaneously with application of the stimulating near-ultraviolet light. The decay with time of the stored energy in the two phosphors is compared.

THE energy storage properties of some silver activated alkali halides have been discussed in several recent publications.^{1,2} The procedure outlined in references 1 and 2 has been to irradiate an excited phosphor with long wave light and observe in a phototube (RCA-1P28) what may be described as a "post-stimulation phosphorescence" of the ultraviolet emission band after the stimulating long wave light has been extinguished. In using the 1P28 as a detector, difficulties are encountered if attempts are made to measure the stimulated emission while the stimulating light is on, because the phototube's spectral response is such as to respond to the stimulating light as well. The stimulated light emitted while the stimulating light is on might be called "co-stimulation phosphorescence." To avoid this problem, the writers have employed photosensitive Geiger counters to detect the stimulated emission. It has long been known³ that photosensitive Geiger counters can be produced which have an excellent sensitivity at 2500A but no response to near-ultraviolet or visible radiations. Accordingly, in the present investigation, photosensitive Geiger counters have been employed to detect the photostimulated emission of the ultraviolet bands of KCl-Ag and NaCl-Ag, the ultraviolet band of KCl-Ag being centered at 2800A and that of NaCl-Ag at 2500A. Irradiation, storage, and measurements relating to all phosphor samples were carried out at room temperature (25°C).

To study the photostimulated emission from NaCl-Ag

and KCl-Ag, a polycrystalline mass of KCl-Ag (AgCl concentration 0.10 ± 0.02 percent by weight) and a single crystal of NaCl-Ag prepared by The Harshaw Chemical Company (AgCl concentration 0.37 ± 0.06 percent by weight) were irradiated by x-rays of maximum energy 25 kev for ten minutes to receive a dosage of three roentgens. The irradiations were carried out in total darkness, and the materials were stored for twenty-four hours in light-tight containers. At the end of that time, the crystals were each stimulated by a one-watt tungsten lamp at a distance of seven centimeters for a period of one minute. The counting rates of NaCl-Ag and KCl-Ag before, during, and after the one-minute period of photostimulation are shown in Fig. 1. A time of one minute before the stimulating light was turned on was taken arbitrarily as time zero. Prior to stimulation, the slow normal unphotostimulated phosphorescence of NaCl-Ag was ~ 35 counts per minute, rising immediately to $\sim 50\,000$ counts per minute in the form of co-stimulation phosphorescence. After one minute of photostimulation, the stimulating tungsten lamp was extinguished, and the luminescence from NaCl-Ag dropped immediately to a post-stimulation phosphorescence count of about 9000 per minute. Thus, photostimulation with the one-watt bulb of NaCl-Ag twenty-four hours after receipt of a dosage of three roentgens gave rise to a co-stimulation phosphorescence 1400 times greater than the residual unphotostimulated phosphorescence (35 counts per minute) existing prior to stimulation, and to a post-stimulation phosphorescence greater than the same quantity by a factor of 257. The similarly exposed KCl-Ag gave no evidence of slow unphotostimulated phosphorescence at 25°C, twenty-four hours after excitation. The counting rate in the photosensitive Geiger counter was only the natural background count. However, upon photostimulation, the

* Assisted by the joint program of the U. S. Office of Naval Research and the U. S. Atomic Energy Commission.

¹ M. Furst and H. G. Kallmann, *Phys. Rev.* **82**, 964 (1951); **83**, 674 (1951); Kallmann, Furst, and Sidran, *Nucleonics* **10**, No. 9, 15 (1952).

² Bittman, Furst, and Kallmann, *Phys. Rev.* **87**, 83 (1952).

³ P. B. Weisz, *Electronics* **19**, No. 7, 106 (1946); H. Friedman and C. P. Glover, *Nucleonics* **10**, No. 6, 24 (1952); C. E. Mandeville and H. O. Albrecht, *Phys. Rev.* **79**, 1010 (1950).