Precision Evaluation of the High-Frequency Limit of the Continuous X-Ray Spectrum Using a Gas Target X-Ray Tube*†

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The major uncertainty in previous measurements of the high-frequency limit of the continuous x-ray spectrum was due to the fine structure associated with the observed cutoff. The development of a high-power mercury vapor target x-ray tube permitted observations to be made under idealized thin-target conditions. The x-ray intensity near the high-frequency limit was comparable to that of a solid target x-ray tube. The x-ray intensity was measured as a function of the voltage applied across the tube, with a double-crystal monochromator adjusted for the 1.537400 kxu wavelength of the Cu $K\alpha_1$ line. The absence of fine structure in the observed isochromat, in contrast to most solid targets, greatly increased the accuracy in the determination of the voltage cutoff. Accurate measurements of this voltage yielded a voltage-wavelength conversion factor $V\lambda_s = (12372.26 \pm 23$ ppm) xu-volts. Using h/e from non-x-ray data, the x-ray wavelength conversion factor from x units to cm (or angstroms) can be calculated, or conversely h/e evaluated. The effects of a revision in the Cu $K\alpha_1$ wavelength, which is now in publication by one of the authors, are also discussed.

INTRODUCTION

ELEMENTARY quantum theory considerations give the short wavelength limit of the continuous x-ray spectrum in terms of the voltage applied to the x-ray tube. This relation is $Ve = h\nu$, which may be rewritten as

$$h/e = V\lambda_s(\Lambda/c^2), \tag{1}$$

where h/e (the ratio of Planck's constant to the charge on the electron) is expressed in erg·sec/esu, V in volts, λ_s in kxu, and c, the velocity of light, in cm/sec. $\Lambda = \lambda_g/\lambda_s$, the x-ray wavelength conversion factor, represents the ratio between wavelength on the Siegbahn scale and wavelength on the absolute scale, i.e., Å/kxu. (1 kxu $= 10^3$ xu.)

This relationship has traditionally been used as a means of measuring h/e, but of course the x-ray measurement only evaluates $V\lambda_s$. The three[1–3] most precise evaluations of $V\lambda_s$ and the calculated values of h/e have yielded values which were considerably lower than those obtained through non-x-ray measurements.[4] The discrepancy was so great that DuMond and Cohen[5] gave such values a low weight in their evaluation of atomic constants while Bearden and Thomsen[4] excluded these measurements entirely. This discrepancy was largely attributed to the fine structure observed at the high-frequency limit of the continuous spectrum. Detailed measurements of the structure as a function of atomic number and voltage applied to the x-ray tube have been made by Johnson[6] and Per Johansson.[7] Its origin has been interpreted by Ulmer[8] as due to solid-state band structure. The initial state of the electron is determined by the cathode of the x-ray tube while the final state is characterized by the target. The distribution of those unoccupied energy levels to which a significant number of transitions are allowed then determines the shape of the continuous spectrum near cutoff. In principle this result holds only for a "thin target"; however since characteristic energy losses[9] occur in discrete steps of the order of 10 eV, it will also hold for a thick target within a few electron volts of the cutoff point.

In order to eliminate the fine structure, due to the solid state, a gas target x-ray tube[10] has been developed in which an electron beam of 20–50 mA and 8–20 kV is focused to a mercury jet as target. The intensity observed near the high-frequency limit is comparable to that of a solid target. This effectively thin target satisfies the assumptions of the Sommerfeld[11] theory and hence greatly simplifies the interpretation of the measurements which can then be used to obtain a refined value of h/e. On the other hand, if h/e is known with sufficiently high accuracy from non-x-ray sources, it gives a means of determining Λ, as is evident from Eq. (1). At the present moment, the relative uncertainty in the value of Λ appears somewhat greater than

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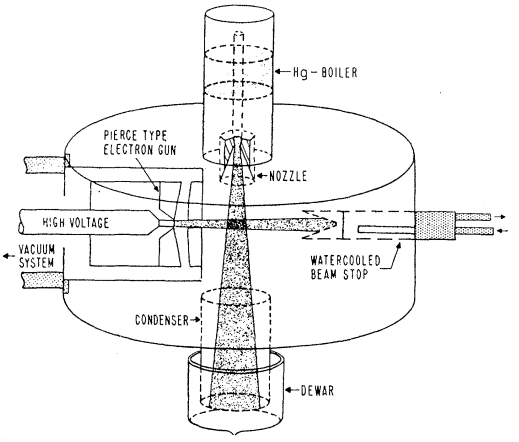

FIG. 1. Three-dimensional view of the gas target x-ray tube.

that of h/e, although both are of the same order of magnitude.

In interpreting the results, certain small corrections to the measured potential are essential, including those due to space charge and to work functions of the cathode and anode.[12] These are obtained, in part by a measurement of the ionization potential of the gas.

GAS TARGET X-RAY TUBE

Two gas target x-ray tubes have been used in the present work. The basic principles of both tubes are shown schematically in Fig. 1. The x rays were observed along an axis which was perpendicular to both the electron beam and the mercury jet.

The required area density of the jet can be estimated as follows: From solid target measurements[13,14] and electron energy losses in thin foils[9] it can be shown that the radiation near the high-frequency limit is produced by a solid target thickness of the order of 500 Å. Thus, to obtain sufficient x-ray intensity for precision measurements, the total atomic cross section for the gas target should be equivalent to a 500 Å thickness of a solid. This was achieved by using a vapor jet formed by a nozzle, as shown in Fig. 1. The diameter of the mercury beam as indicated by the focal spot was approximately 5 mm. The vapor was formed by an electrically heated Hg boiler, and the jetstream directed into a dry-ice cold trap. The freezing out of the Hg vapor greatly reduced the pumping speed required to maintain a pressure of 10^{-4} mm of Hg in the target chamber. A 500 liters/sec mercury vapor diffusion pump and liquid nitrogen traps were used to evacuate the electron gun and high voltage section of the x-ray tube.

The electron beam was formed by a two-element Pierce electron gun,[15] with a unipotential dispenser cathode.[16] This gun operated very satisfactorily, and at 8 kV produced a maximum beam current of 50 mA with a beam diameter of about 3 mm. The accelerating anode of the Pierce gun, the electron collector (beam stop), the nozzle, and the condenser were all electrically connected and maintained at ground potential, except for possible small effects such as contact and thermal emf's. The presence of Hg atoms in the tube largely neutralized the space charge[17] and collimated the electron beam through gas focusing. However, the ion bombardment of the cathode reduced its life to approximately 40 h.

In order to obtain effective work function corrections, as described in the section on corrections and probable errors, the ionization potential of Hg was measured by the usual method. This required a shielding grid at the potential of the target chamber and an ion collecting plate at a small negative potential. (These auxiliary electrodes are not shown in Fig. 1.) In the first tube a small grid and plate were inserted through the port where the x-ray beam was normally observed. In the second tube, both the stainless steel grid and plate were cylindrical and concentric with the mercury jet, with openings to permit passage of the electron and x-ray beams; they remained in place throughout the x-ray measurements, permitting an ionization curve to be run after each isochromat.

The first tube[18] was essentially two intersecting stainless steel cylinders each 2 in. in internal diameter and 4.5 in. long. The mercury jet was directly vertically upwards and an arrangement provided for condensing the mercury and returning it to the boiler. The second tube was designed on a larger scale, the actual geometry being very similar to the schematic shown in Fig. 1. The enclosing steel cylinder was 11 in. i.d. and 3.5 in. in height, with end plates of 0.5 in. thickness. The mercury jet was directly vertically downwards; no return was provided, since less than 300 cc of mercury was required for a 12-h run.

POWER SUPPLY AND MONOCHROMATOR

Power supply. The high-voltage regulated power supply and the high-voltage measurement circuit were basically the same as reported by Bearden and Schwarz,[1] with minor modifications. The regulating amplifier was replaced by a commercial high-gain operational amplifier, and a phase-shift-free voltage divider[19] was designed for the feedback loop. This circuit reduced the ripple voltage to 0.05 V peak to peak.

The high voltage V_m (see Table I) was measured with

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TABLE I. Measurements for the voltage-wavelength conversion factor.

	Run 1	2	3	4
		High-voltage measurement		
E (St. cell)	1.018 414	1.018 414	1.018 414	1.018 414
ΔE ($K-3$ pot.)	0.018 256	0.018 191	0.018 231	0.018 278
$E-\Delta E$	1.000 158	1.000 223	1.000 183	1.000 136
V_m	8048.96	8049.48	8049.16	8048.78
		Corrections		
V_i	10.434	10.434	10.434	10.434
V_{mi}	10.32	10.57	10.57	10.05
V_s-V_{si}	0.90	0.92	0.83	0.83
V_D	0.07	0.07	0.07	0.07
V	8048.10	8048.35	8048.12	8048.26
		Wavelength		
λ_s(xu)	1537.400	1537.370	1537.395	1537.403
		Voltage-wavelength conversion factor		
$V\lambda_s$[a]	12 373.14	12 373.29	12 373.13	12 373.41
Weight	1	2	2	2

[a] Weighted average $V\lambda_s = (12\,373.26 \pm 23$ ppm) xu·V.

a precision voltage divider and a potentiometer. The precision voltage divider, consisting of an 8526-Ω protective resistor in series with two 1-MΩ standard resistors (each with 10 subdivisions) and two standard resistors of 100 Ω, was so arranged that the smaller voltage was almost exactly equal to that of a standard cell for the average value of high voltage used in the measurements. The precision potentiometer then was used only to measure the small incremental difference between the voltage drop in the two 100-Ω standards of the divider and that of the standard cell. Hence the high voltage V_m was determined from the difference between the emf of the NBS 968 saturated standard cell and the potentiometer reading ΔE, multiplied by the voltage divider ratio of 8047.688. The standard resistors and standard cells used have been calibrated by the National Bureau of Standards a number of times, including one check during the present experiment.

Monochromator. A double-crystal spectrometer,[20] with quartz crystals cut parallel to the $10\bar{1}1$ plane, was used for a monochromator with a resolution of approximately 13 000 as determined from the parallel (1, −1) position rocking curve. The monochromator was adjusted for the wavelength of the Cu $K\alpha_1$ line, which is one of the best known standard x-ray wavelengths. The monochromator was aligned by replacing the vapor target with a Cu target, and accurately calibrated against the $K\alpha_1$ line, assuming a peak wavelength of 1.537400 kxu.[21] In the different experimental runs the exact wavelength setting was determined with respect to this peak angle position. The x rays were detected by a scintillation counter with a 25-mil-thick NaI(Tl) crystal placed on a selected photomultiplier tube and shielded by a low radioactive iron housing. For a pulse-height discriminator setting which gave 80% detection efficiency the background was 0.3 counts/min.

ISOCHROMATS

In principle the high-frequency limit of the continuous spectrum can be found most simply by holding a fixed voltage on the x-ray tube and determining the shortest wavelength emitted. In practice it is usually more convenient to adjust the monochromator for a fixed wavelength and determine the variation of intensity as the voltage is changed in small steps. The resulting curve is called an isochromat.

Four averaged isochromats were used in the four runs shown in Table I. These were obtained as follows: Each averaged isochromat is the composite of ten individual isochromats. Seventeen voltage points in the vicinity of cutoff were selected and the x-ray intensity recorded for a period of 200 sec at each point. In order to minimize instrumental drift, these measurements were first recorded for increasing voltage and then repeated for decreasing voltage for each individual isochromat. After initial adjustment of the apparatus was completed and definitive recording was begun, no individual isochromat was rejected for any reason. One of the averaged isochromats is indicated by the data points in Fig. 2, where the normalized intensity is plotted against voltage change. Measurements up to 500 V above the short wavelength limit indicated no further appreciable increase in intensity.

Since the profile near the high-frequency limit is of primary interest, both for an h/e determination (or $V\lambda_s$) and from a theoretical standpoint, the observed

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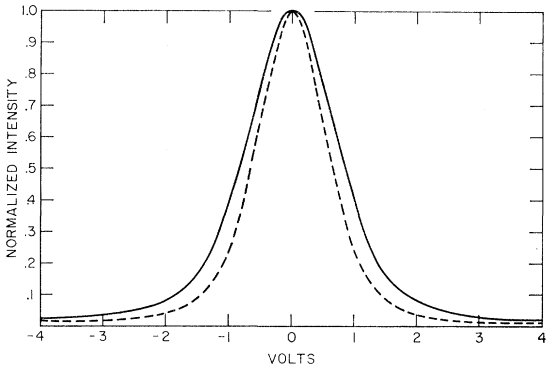

FIG. 2. Experimental window (solid line) and theoretical window (dashed line) of the double-crystal monochromator.

isochromat was corrected for instrumental smearing, which is primarily due to the width of the monochromator window. This window, shown in Fig. 3, was obtained from the parallel position (1,−1) rocking curve using a copper target. The double crystal diffraction pattern was also calculated from the Darwin-Prins-Ewald theory of single crystal diffraction.[22] The greater width of the observed pattern must be attributed to slight imperfections in the quartz crystals. A study of the crystals[23] showed that they were not perfect specimens. Since a 3 cm width of the crystal was used for both copper and vapor targets (due to the extended x-ray source), the crystal irregularities broadened the diffraction pattern.

The measured window was used to correct the observed isochromat, but the extended wings made an unfolding procedure inaccurate. This difficulty was avoided by using an IBM-7094 computer to fold assumed isochromat profiles with the observed window and finding the best fit for the observed data. The assumed profiles were based on the Sommerfeld theory,[24] which gives the x-ray intensity as a constant independent of the frequency for $h\nu<eV$, and zero for $h\nu>eV$. However, near cutoff the shape of the profile is determined by the distribution of final states available to the electron. If one assumes no electric or magnetic field at the target, the density of final states is proportional to $(\Delta E)^{1/2}$, where $\Delta E=e(V-V_0)$ or the energy difference between the incoming electron and the emitted photon.[25]

Consequently the x-ray intensity was represented by the empirical relation

$$I=I_0\{1-\exp[-\alpha(V-V_0)^{1/2}]\}, \qquad (2)$$

where α is an adjustable parameter. Equation (2) approximates the Sommerfeld profile, except near the high-frequency limit, and becomes proportional to the density of the final states as this limit is approached. The observed x-ray intensity should be given by folding Eq. (2) with the observed window of Fig. 3.

The parameters V_0 and α were varied stepwise; for each pair of values an isochromat was computed, and compared with the observed curve. The best fits were obtained with values of α between 1 and 2 V^{-1}; outside this range the predicted isochromat varied significantly from the observed one. The isochromats for these two values of α are shown in Fig. 2. For the best fit the value of V_0 differed only about 5 ppm from the voltage at the half-intensity point; hence, the latter was used as a sufficiently accurate criterion for the high-frequency cutoff.

CORRECTIONS AND PROBABLE ERRORS

Work Function Correction

The voltage measured, V_m, represents the potential across the x-ray tube and not the electron energy, since contact potentials and the initial thermal electron energy must be taken into account. In the solid target h/e experiments, the applied potential must be corrected for the work function of the cathode only; the work function of the anode is not involved, since the electron comes to rest near the Fermi level of the anode. For the gas target tube, however, both initial and final states are those of a free electron, and the difference between the work function of the cathode φ_c and the anode φ_a plus the thermal energy kT must be added to the measured potential.

The retardation of the electrons due to the space charge *at the cathode* has the effect of increasing the cathode work function. This space charge potential V_s

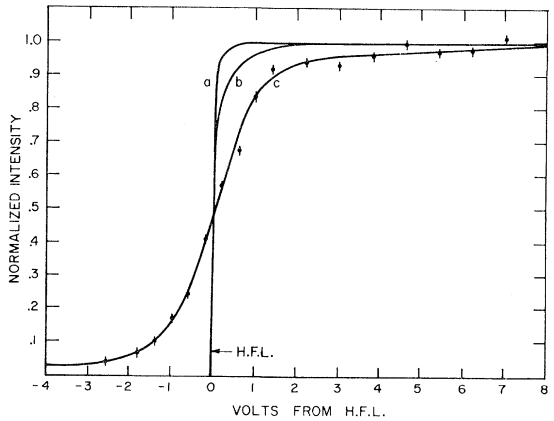

FIG. 3. (a) Assumed isochromat, Eq. (2), with $\alpha=1$ V^{-1}; (b) assumed isochromat with $\alpha=2$ V^{-1}; (c) predicted isochromat obtained by folding either assumed curve with the experimental window of Fig. 2; the predicted curves based on (a) and (b) showed no significant difference. Experimental data points are shown as dots, with bars indicating the statistical errors.

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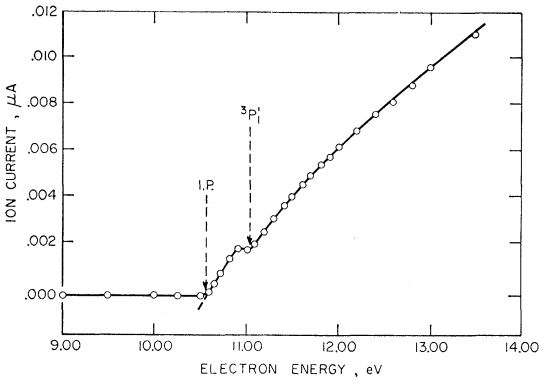

FIG. 4. Experimentally determined probability of ionization by electron impact in Hg gas. Cathode temperature 1493°K.

can be calculated[26] from the cathode work function φ_c, the critical cathode temperature θ (where the space charge minimum disappears), and operating temperature T, by

$$V_s=\varphi_c\left(\frac{T-\theta}{\theta}\right)-\frac{kT}{e}\ln\frac{I}{I_\theta}. \quad (3)$$

In this expression I_θ represents the cathode current at the critical temperature θ, and I that at operating temperature T, while k and e are the Boltzmann constant and the electronic charge. With the above-mentioned corrections to the measured voltage V_m, the effective difference V becomes

$$V=V_m+V_{kT}+(\varphi_c-\varphi_a)+V_s. \quad (4)$$

To determine these correction terms experimentally, a measurement was made of the ionization potential[27] of Hg (for which a value of 10.434 V is known from spectroscopic data). The observed probability of ionization by electron impact for Hg vapor is indicated in Fig. 4. The structure in the curve is due to auto-ionization[28] corresponding to the $^3P_1{}^1$ state of mercury 0.6 eV above the ground state, which competes with the ionization process.

For the ionization potential of mercury at the same cathode temperature, the relation corresponding to Eq. (4) is

$$V_i=V_{mi}+V_{kT}+(\varphi_c-\varphi_a)+V_{si}, \quad (5)$$

where the subscript i denotes quantities involved in the ionization potential measurement. Subtracting Eq. (5) from Eq. (4) and making use of Eq. (3) now yields

$$V=V_m+V_i-V_{mi}-\frac{kT}{e}\ln\frac{I}{I_i}, \quad (6)$$

the last term being simply V_s-V_{si}. Numerical values of these correction terms are shown in Table I.

Potential Depression

Since there is no external electric field acting on the electron beam in the target chamber, the space charge effects could produce a potential variation across the electron beam, which might become significant at moderate current densities.[29] The space charge effect would lower the electron energy. It should also produce an energy spread of several electron volts in the cutoff of the isochromat, unless it is largely neutralized by the presence of positive ions, which are trapped by the electron beam. Linder and Hernqvist[30] have studied the degree of neutralization of space charge by positive ions, and derived an expression for the potential depression as a function of the gas pressure.

For an operating pressure of 10^{-5} mm Hg in the described apparatus, the calculated voltage depression V_D is 0.07 V (Table I). This could be in error by 0.07 V, since complete neutralization would take place at 2×10^{-4} mm Hg and the pressure measurement could be in error by an order of magnitude.

Errors

The probable errors, in parts per million (ppm) of the final result are estimated as follows:

1. Determination of the voltage at the isochromat cutoff:	
Potentiometer measurements	3.5 ppm
Precision voltage divider	2.0
Isochromat cutoff	12
Work function correction	13
Voltage depression	9
Total (rms)	21 ppm
2. Spectrometer calibration	10 ppm

Total probable error in the voltage-wavelength conversion factor is thus 23 ppm; the error in the conversion factor from NBS to absolute volts (5 ppm) does not significantly increase this figure.

The four separate runs were assigned weights based on the statistics of the isochromat recording.

RESULTS

A summary of the experimental results is given in Table I. The total recording time for each run was slightly less than 19 h. Runs 1 and 2 were made with the tube briefly described in the gas target x-ray tube section and reported in detail in the thesis of one of the authors (J. J. S.). Runs 3 and 4 were made with the tube design shown in Fig. 1. The excellent agreement of the final values lends support to the methods and experimental measurements involved in applying the necessary corrections.

[26] Wayne B. Nottingham, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 21, p. 42.
[27] S. C. Brown, *Basic Data of Plasma Physics* (John Wiley & Sons, Inc., New York, 1959), p. 14.
[28] W. M. Hickam, Phys. Rev. **95**, 704 (1954).
[29] O. Klemperer, *Electron Optics* (Cambridge University Press, London, 1953), p. 210.
[30] E. G. Linder and K. G. Hernqvist, J. Appl. Phys. **21**, 1088 (1950).

The voltage of standard cell E (actually three were used and frequently intercompared) has been corrected for the difference between the National Bureau of Standards volt and the absolute volt, using the conversion factor given by Driscoll and Cutkosky[31]

$$1 \text{ NBS volt} = 1.000\,010 \text{ absolute volts} \pm 5 \text{ ppm}.$$

ΔE is the incremental voltage measured by the K3 potentiometer.

As shown in Table I the weighted average of the above measurements gives

$$V\lambda_s = (12\,373.26 \pm 23 \text{ ppm}) \text{ xu-volts}.$$

Combining this with the commonly accepted value of $\Lambda = 1.002\,02 \pm 20$ ppm and $c = 299\,792.5$ km/sec,[32] one obtains

$$h/e = (1.379\,49 \pm 30 \text{ ppm}) \times 10^{-17} \text{ erg} \cdot \text{sec/esu}.$$

The non-x-ray value[32] is

$$h/e = (1.379\,47 \pm 7 \text{ ppm}) \times 10^{-17} \text{ erg} \cdot \text{sec/esu}.$$

The probable errors overlap, and for the first time the x-ray and non-x-ray values of h/e are in good agreement. However, the probable error of the x-ray value is more than four times that of the non-x-ray one. Approximately half of this is due to the uncertainty in the value of Λ.

DuMond and Cohen[33] have pointed out that the optical calibration lines used by Tyrén[34] in his concave grating measurement of Λ require correction for the Lamb shift. This might increase his reported value of $\Lambda = 1.001\,99$ by the order of 50 ppm (due to loss of data and plates it is impossible to determine the exact correction). DuMond and Kirkpatrick in a recent report to the National Science Foundation on a new concave grating experiment give a provisional value

$$\Lambda = 1.001\,80 \pm 10 \text{ ppm}.$$

This is about 200 ppm lower than Tyrén's value or the plane grating values. Thus the true value of Λ may be considerably outside the $1.002\,03 \pm 20$ ppm range calculated by Birge,[35] based principally on measurements with plane gratings.

From these considerations the measured value of $V\lambda_s$ could best be used to evaluate Λ. With $h/e = (1.37947 \pm 7 \text{ ppm}) \times 10^{-17}$ erg·sec/esu[32] the value is

$$\Lambda = 1.002\,005 \pm 24 \text{ ppm}.$$

This value is based on Cu $K\alpha_1 = 1537.400$ xu. However, a report by one of the authors (J. A. B.)[36] on a serious wavelength discrepancy of approximately 20 ppm between the Mo $K\alpha_1$ and the Cu $K\alpha_1$ xu standards has been confirmed and is in preparation for publication. On the basis of Mo $K\alpha_1$ as 707.831 xu (which is taken as a provisional definition of the xu in this report[37]) the wavelength of the Cu $K\alpha_1$ is 1537.370 xu. With this revision the experimental value becomes

$$V\lambda_s = (12\,373.02 \pm 23 \text{ ppm}) \text{ xu} \cdot \text{V}$$

and

$$\Lambda = 1.002\,024 \pm 24 \text{ ppm}.$$

(This revision will not significantly alter the value of h/e calculated above, since the change in Cu $K\alpha_1$ wavelength also implies a compensating change in the ruled grating values of Λ.)

CONCLUSION

The absence of structure and the sharpness of the high-frequency limit of the continuous x-ray spectrum, together with a measurement of the work function correction made possible a precision determination of $V\lambda_s$. The lack of structure in the gas target isochromat tends to support Ulmer's theory as to the origin of the structure. The profile of the isochromat gave the shape of the continuous spectrum produced by single electron bombardment. The measured intensity was constant from a few eV of cutoff to 500 eV above in accordance with the Sommerfeld theory.

With the use of the non-x-ray value of h/e, a precision determination of Λ has been obtained. The accuracy of this value appears comparable to those obtained by either ruled-grating or crystal measurements.

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