Measurement of Absolute X-Ray Intensities and Absolute Sensitivity of X-Ray Film with a Geiger-Müller Counter*

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A special Geiger-Müller counter for measuring absolute intensities of x-rays of known wave-lengths is described. The rays pass through the counter in such a way that only electrons set free in the gas of the counter are counted. The absorbing gas column is krypton at 6.31 cm pressure, 1.08 cm thick, and 0.0062 cm² in cross section. For fluorescent Zr K-rays, the absorption in the counter is 3.75 percent, as computed on the basis of the energy distribution of the radiation: $K\alpha : K\beta : K\gamma = 0.829 : 0.154 : 0.017$, and absorption coefficients computed from Richtmyer and Warburton's formulae. A "standard beam" of fluorescent Zr-rays from a specially constructed x-ray tube has been calibrated with the counter. With this beam, the minimum exposure of x-ray film for detectable blackening is found to be about 0.72×10^6 quanta cm^{-2}, incident on the film, or 4.1×10^4 quanta cm^{-2} absorbed in the emulsions (Eastman Ultra-Speed Duplitized X-Ray Film). The total absorption coefficients of the film, celluloid, and emulsion, for the Zr-rays, are found to be, respectively, 8.63 cm^{-1}, 1.66 cm^{-1} and 13.0 cm^{-1}. Time-blackening curves are given. These show discontinuities at exposures of about 10^7 quanta cm^{-2}.

I. Introduction

The great sensitivity of Geiger-Müller counters for counting charged particles is well known. Besides their applications to problems of cosmic rays and radioactivity, in which the electrified particles penetrate the counting tubes from outside, they have been adapted for counting photoelectrons liberated internally by light, γ-rays and x-rays. The work reported here concerns the use of a counter for measurements of feeble x-ray intensities, by means of the photoelectric action of the x-rays on the gas of the counter. By this method it is possible to make absolute measurements of the flux of x-radiation with as good accuracy as that with which the energy-distribution of the incident radiation and its absorption coefficients in the gas are known.

Huppertsburg[1] has employed a counter with solid metal walls for comparison of the intensities of heterogeneous x-rays, and deduced the absolute sensitivity of the counter by measuring the corresponding ionization produced in an ionization chamber whose sensitivity he calculated. He showed that the counting rate was proportional to the area exposed, for a given x-ray beam, and that the photoelectrons came almost exclusively from the walls of his counter; the absorption in the gas was negligible.

In the experiments described here, almost monochromatic fluorescent x-rays are used. Absolute intensities are measured by means of the photoelectric action of the rays on the *gas* of the counter, by measuring the counting rates, using the efficiency of the counter as computed from the energy-distribution of the incident rays, the absorption coefficients of the gas, and the thickness and pressure of the gas. The counter thus forms a small ionization chamber in which the number of individual photoelectrons is counted, instead of measuring the amount of ionization in the customary manner.

II. The Counter

Figs. 1, (a) and (b), are respectively, an x-ray photograph of the counter and a photograph of its cylinder-electrode (cathode). The x-rays enter and leave the counter through "bubble" windows of very thin glass, concave inwards. A narrow pencil of rays passes through the counter along a transverse diameter of the cylinder, in such a way as to miss the wire electrode, which is slightly off center. The cathode is a heavy gold-plated brass cylinder, 1.0 cm in diameter and 1.8 cm long, slightly flared at the edges. Two small lead blocks inserted in its walls collimate a beam of rays 0.0062

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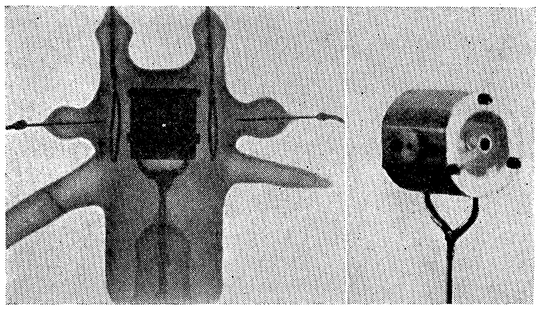
[1] A. Huppertsburg, Zeits. f. Physik **75**, 231 (1932).



a b

FIG. 1. X-ray photographs of apparatus.

cm^2 in cross section by means of holes, respectively, 0.089 cm and 0.225 cm in diameter, for the entrance and exit of the rays. The inner surfaces of the windows are covered with cellophane disks, 0.0025 cm thick; these are sufficient to exclude the entrance of electrons liberated elsewhere than inside the cylinder, but their total absorption is so small that the scattering and photoelectron emission from the cellophane itself may be neglected. The ends of the cylinder are covered with disks of thick mica, to prevent stray ions from diffusing into the sensitive region.[2] The wire electrode is of bare tungsten, 0.0076 cm in diameter. The gas consists of approximately 98.5 percent krypton and 1.5 percent xenon at 6.31 cm of mercury, at 27.35°C.

Externally, the counter is shielded from stray radiation by sheets of lead so arranged that the number of accidental impulses due to stray radiation is reduced to a (measured) minimum. A series of thick lead blocks with holes of suitable diameters, mounted between the x-ray source and the counting tube, restrict the incident beam of x-rays to a size only a little greater than that allowed to pass through the counter.

The counter must be very accurately aligned with the source of x-rays, in order that the area of the beam passing through it shall be the same as that of the front collimating hole of its cathode, and in order that none of the radiation passing through the cathode shall strike the metal surface. This is accomplished by sighting through the small holes of the counter on the source of x-rays, which is illuminated with a bright light, with a telescope placed several meters beyond the apparatus.

Apart from the loss of impulses due to plurality of discharges falling within intervals less than the resolving-time of the apparatus, and the spurious discharges due to radiations of various kinds, there is assumed to be a one-to-one correspondence between the number of photoelectric absorption events in the counter and the number of impulses recorded; i.e., the efficiency of the counter for counting the photoelectrons liberated in it is taken as $1-\epsilon$, where ϵ is the loss due to multiple discharges occurring within the resolving-time of the apparatus. This seems reasonable, since each photoelectron produces some 700 pairs of ions, any pair of which would suffice to discharge the counter.

III. Amplifying and Recording Apparatus

The circuit used for amplifying and recording the counter impulses has been described elsewhere.[3] It consists of a two-stage resistance-coupled amplifier with high-gain tubes (RCA 257), driving a pair of grid-controlled gaseous-discharge tubes (W.E. 256–A), arranged in a Wynn-Williams thyratron circuit. The significant time constants of the apparatus were found by oscillograph measurements to be approximately as follows: (a) recovery time of the counter and its attendant circuit, 0.0038 sec.; (b) resolving time of the circuit with the pair of relay tubes, <0.0007 sec.; (c) resolving time of the electromechanical impulse counter (a watch driven by a light relay), with one pair of relay tubes, ~ 0.008 sec., or 0.004 sec., with two pairs, and so on. The high-voltage supply is obtained from a rectifier of the type designed by Street and Johnson,[4] and is constant to <0.1 percent.

The counter wire is coupled to the amplifier through a $25\mu\mu$f condenser, and to ground through a resistance of 3.3×10^8 ohms.

Correction for loss of impulses, due to the arrival of more than one impulse within the resolving time of the apparatus, may be made by

[2] The two loop electrodes, shown in Fig. 1a, were inserted for collecting stray electrons, but were not used.

[3] G. L. Locher, J. Frank. Inst. **216**, 553 (1933). Telephones are inserted at J (Fig. 1).

[4] J. C. Street and T. H. Johnson, J. Frank. Inst. **214**, 155 (1932). The "first type" of voltage control is used. The tube for regulating the voltage is of type 57; this gives better regulation than a 24-A tube.

means of the formula $N_{\text{true}}=N_{\text{observed}}(x/1-e^{-x})$, where x is the average number of impulses arriving within the longest resolving time of the apparatus.[3] This correction is precise if the resolving time is precisely known. In this experiment, however, the rates of counting have mostly been kept sufficiently low that the loss due to this effect may be neglected. By using a low counting rate, one also avoids the danger of error due to change of apparent sensitivity of the counter with high speeds of operation.[5]

IV. Source of X-Rays

Fig. 2 shows the x-ray tube, which has a molybdenum anode and a zirconium metal fluorescer, F, arranged inside the tube in a region of intense radiation density. The fluorescent radiation emerges through a very thin window, W, of Pyrex glass. X-rays from about 1.06 cm^2 area of the fluorescer pass through holes in a series of lead baffles into the counting tube.

The incident radiation is assumed to be purely fluorescent Zr K-radiation, with wave-lengths and energy distribution as given in Table I.

V. Quantum Efficiency of the Counter

The fraction of the x-rays absorbed in the gas is calculated on the basis of the wave-lengths and energy-distribution given above, the pressure and constitution of the gas, and the length of path in the counter. In krypton and xenon, the scattering may be neglected, as compared with the fluorescent absorption. The atomic fluorescent absorption coefficients, τ_a, of the gas are calculated from the formula of Richtmyer and Warburton,[6] in Table II.

The number of atoms per cc in the gas (98.5 percent Kr+1.5 percent Xe at 6.312 cm mercury, at 27.35°C) is 2.043×10^{18}. The absorbing path in the counter is 1.079 cm.

From the data given above, we get the fractions $f_{K\alpha}$, $f_{K\beta}$, and $f_{K\gamma}$, of the $K\alpha$, $K\beta$, and $K\gamma$ radiation absorbed in the counter, namely,

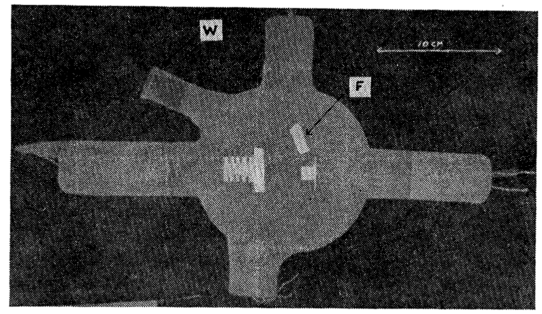


Fig. 2. Photograph of x-ray tube.

Table I. *Energy-distribution of Zr K-radiation.*

Wave-lengths[7]

$K\alpha_1=0.7843$A $\quad K\beta=0.7005$A

$K\alpha_2=0.7885$A $\quad K\gamma=0.6883$A

Weighted mean $K\alpha=(2K\alpha_1+K\alpha_2)/3=0.7857$A

Energy-distribution

$K\alpha/K\beta=5.38$;[8] $\quad K\beta/K\gamma=9.0$;[9]

or $\quad K\alpha : K\beta : K\gamma=48.42 : 9 : 1$

Relative numbers of quanta

$N_{K\alpha} : N_{K\beta} : N_{K\gamma}=0.845 : 0.140 : 0.015$

Table II. *Atomic fluorescent absorption coefficients* $\times 10^{20}$.

	Krypton	Xenon	98.5% Kr+1.5% Xe
$K\alpha_{1,2}$	1.857	1.362	1.818
$K\beta$	1.294	0.965	1.289
$K\gamma$	1.225	0.913	1.220

$$f_{K\alpha}=1-e\exp[-\tau_{K\alpha}x]=0.0393 \text{ or } 3.93 \text{ percent}$$
$$f_{K\beta}=1-e\exp[-\tau_{K\beta}x]=0.0280 \text{ or } 2.80 \text{ percent}$$
$$f_{K\gamma}=1-e\exp[-\tau_{K\gamma}x]=0.0265 \text{ or } 2.65 \text{ percent.}$$

For the mixture of radiation in the beam, we have the total fraction, f, absorbed: $f=0.0375$ or 3.75 percent.

If every photoelectron discharges the counter, the total efficiency is, accordingly, 3.75 percent; and the multiplication factor between the number of quanta passing through the counter and the number of discharges is 26.67. We here assume that the radiation is pure fluorescent Zr

[5] A spurious increase of sensitivity after rapid operation is especially found in some counters containing photosensitive materials (G. L. Locher, Phys. Rev. **42**, 525 (1932); it usually becomes more conspicuous as the voltage applied to the counter is raised.

[6] F. K. Richtmyer and F. W. Warburton, Phys. Rev. **22**, 539 (1923).

[7] A. H. Compton, *X-Rays and Electrons*, p. 396.

[8] Interpolated from data of E. C. Unnewehr, Phys. Rev. **22**, 529 (1923). We assume that the ratio of the areas under the Zr $K\alpha$ and Zr $K\beta$-lines is the same for fluorescent as for characteristic radiation. A. H. Compton found this to be true for silver (Proc. Nat. Acad. Sci. **14**, 549 (1928)).

[9] Interpolated from data of A. E. Lindh, *Hand. der Exp. Physik*, Vol. 24, Part 2.

K-radiation, also that its energy-distribution is not appreciably altered by selective absorption in materials between the fluorescer and the counting chamber.

VI. Calibration of the X-Ray Beam

A "standard" x-ray beam has been calibrated for use in measurements of the sensitivity and blackening of films. Table III gives the data on this beam and illustrates the method by which the counter was used to calibrate it.

Table III. *"Standard" beam of fluorescent Zr x-rays.*

Voltage applied to counter: $680 \pm 0.1\%$
Area of beam traversing counter: 0.0062 cm²
Distance between fluorescer and counter: 58.8 cm
Potential applied to x-ray tube: 29,400 volts, peak, $\pm$ about 5%; constant to about 1%
Current through x-ray tube: 0.003 ampere$\pm$about 1%
R_D = "accidental" counting rate: front aperture of counter covered, x-ray tube running, $=61.8$ min.$^{-1}$ $\pm 1.26\%$ (6300 impulses)
R_L = correction for light from tube filament[10] $=6.5$ min.$^{-1}$ $\pm 7.1\%$ (196 impulses)
R_T = total rate of counter, with apertures open, $=281$ min.$^{-1} \pm 0.58\%$ (29,600 impulses)
R_X = rate due to x-rays, only, $=213.6$ min.$^{-1}\pm$about 1%
N_C = No. quanta passing through counter per minute $=26.67\times 213.6=5696.7$
N_0 = No. quanta cm^{-2} min.$^{-1}=5696/0.0062=9.03\times 10^5$, approx.
N_0' = No. quanta cm^{-2} sec.$^{-1}=1.50\times 10^4$, approx.

VII. Measurements on X-Ray Film

Eastman Ultra-Speed Duplitized X-Ray Film[11] was used in the experiments described below. The exposed films were all developed for 5 min. at 18°C in Eastman D–19 developer; films exposed simultaneously were developed simultaneously. In all exposures, the x-rays were incident on the side of the film which has the heavy emulsion.

The energy flux at the film (57.3 cm from the fluorescer) was approximately 9.57×10^5 quanta cm^{-2} min.$^{-1}$, or 1.60×10^4 cm^{-2} sec.$^{-1}$. The samples of film were held between sheets of opaque paper in a heavy brass cassette which was perforated on both sides for the entrance and exit of the x-rays. The blackening effect of the softer fluorescent rays from the brass appears to have been small. Three kinds of experiments were performed.

(1) Minimum number of quanta for detectable blackening

Films were exposed to the standard beam for a series of short intervals and developed under identical conditions. An exposure of about 45 seconds gave visible blackening; this corresponds to 7.2×10^5 quanta per cm². The sensitivity of this film seems surprisingly high; if the quanta were spread uniformly over the surface, only 85 per mm are required to give visible blackening.

(2) Absorption coefficients of film, emulsion and celluloid

These were determined by measuring the counting rates with the standard beam, filtered, respectively, through 10 thicknesses of film (0.250 cm) and 10 thicknesses of celluloid from which the emulsions had been dissolved away (0.204 cm). The total absorption coefficients found in this manner are: $\mu_{\text{film}}=8.63$ cm^{-1}; $\mu_{\text{celluloid}}=1.66$ cm^{-1}; $\mu_{\text{emulsion}}=13.0$ cm^{-1}. These values are subject to any error arising from modification of the wave-lengths and energy-distribution while passing through the absorbing material. Such error is probably small, since the incident x-rays are almost monochromatic. The results are believed to be reliable to within 10 percent.

Using the coefficients given above, we may calculate the number of quanta absorbed in the emulsion, for detectable blackening. This is approximately 4.1×10^4 cm^{-2}, or 20 quanta per mm, if spaced on a uniform lattice.

(3) Time-blackening experiments

Fig. 3 shows the time-blackening curve for the film, which was exposed to the standard beam for intervals of 2.5 to 60 minutes. The "blackening," as measured with a densitometer, is defined as the difference between the amounts of light transmitted through the background of the film and the exposed spot, respectively, called L_0 and L. These data are also shown in curve C, Fig. 4, in which the "density of blackening," D, is plotted against time. D is defined thus: $D=\log_{10} L_0/L$.

[10] The counter responds to far-violet and ultraviolet light, in spite of the gold plating on its cathode, and the fact that its windows are of Pyrex glass. The transmission of the light through the windows results from their thinness (about 0.002 cm each).

[11] The film was very kindly supplied by the Eastman Kodak Co. for the purpose.

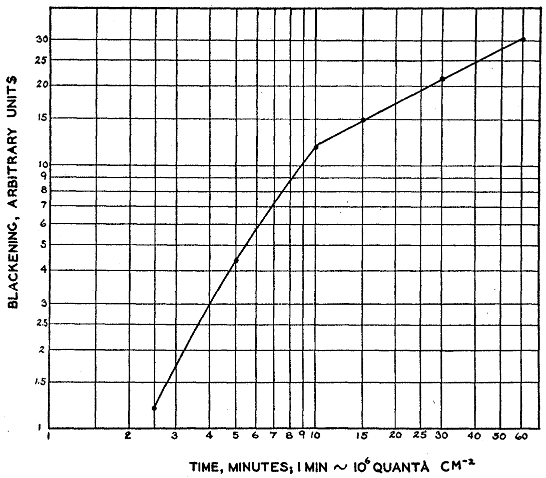


FIG. 3. Blackening of film exposed to x-rays.

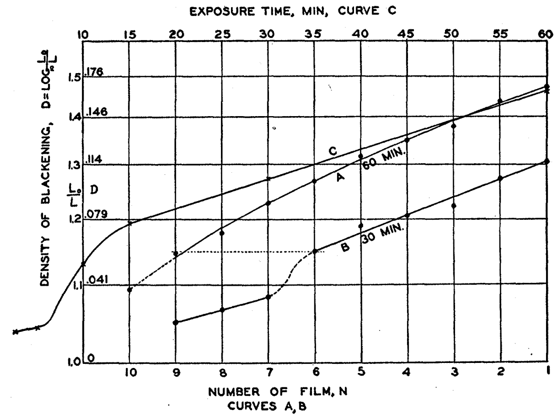


FIG. 4. Blackening of films through which the x-ray beam passed successively.

In another series of exposures, the beam of x-rays passed successively through 10 thicknesses of film, in the manner used by Brindley and Spiers.[12] The blackening of successive films was then measured with a densitometer. Curves A and B, Fig. 4, show the results for exposures of 60 and 30 minutes. The densities of blackening shown on these curves are very much smaller than those given by Brindley and Spiers; the two sets of data may accordingly be regarded as complementary, within the limits of the equivalence of the wave-lengths of the incident x-rays and the unknown equivalence of characteristics of the films used.

The authors wish to express their appreciation to Dr. W. F. G. Swann for his discussions of the problem and his interest in the investigation.

[12] G. W. Brindley and F. W. Spiers, Phil. Mag. **16**, 686 (1933).

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A Contribution to the Theory of Barrier Layer Cells

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It is shown that the theory of the photovoltaic effect developed by Frenkel and Joffe is incomplete because it contains no adequate explanation of the direction of flow of the current. The proper direction of current flow follows as a natural consequence if the great difference in rate of energy loss by photoelectrons in the semiconductor and the metal is taken into account. In the metal collisions between photoelectrons and conduction electrons are frequent, and transfer of energy is rapid, whereas in the semiconductor such collisions are infrequent, and transfer of energy by elastic collisions of a photoelectron with atoms is slow. Hence rapidly moving photoelectrons tend to migrate into the metal and there lose their energy and become trapped.

INTRODUCTION

A BARRIER layer photo-cell such as the Cu_2O or Se cell consists of a closed conducting circuit which comprises a semiconductor forming a link in a metallic circuit, and a very thin barrier to the passage of electrons between the metal and semiconductor at one junction. In order for the cell to function, ionization must be produced in the semiconductor close to the barrier layer. This causes a continuous flow of electrons across the barrier layer from the semi-

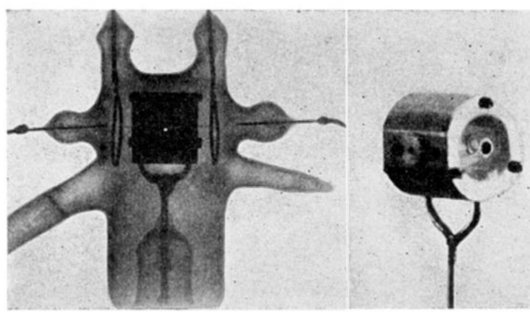

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FIG. 1. X-ray photographs of apparatus.

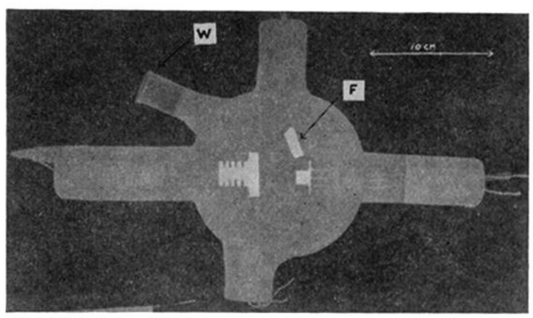

FIG. 2. Photograph of x-ray tube.