THE DETERMINATION OF ABSOLUTE PHOTON FLUXES AND APPLICATIONS TO LABORATORY CALIBRATION PROCEDURES IN THE 100A TO 300A RANGE

by D. H. Tomboulian

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SUMMARY

The problems and methods of making absolute measurements of the intensity of photon fluxes in the wavelength region from 100 to 300 angstroms are discussed. It is shown that a modified Geiger-Mueller counter can provide a solution to this problem. The actual application of such a Geiger-Mueller counter to the determination of the absolute photosensitivity of a specially developed open-window photomultiplier tube and of two different photographic emulsions is described. The experimental photosensitivities determined by these calibrations are also given.
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INTRODUCTION

The problem of making absolute intensity measurements of photon fluxes has recently received emphasis because of increased interest in spectral studies in the vacuum-ultraviolet and in the soft x-ray region. The term absolute will be used to characterize an intensity measurement which is expressible in units of power per unit area or in terms of photon fluxes. Need for such measurements arises when, as in solar spectrophotometry, one wishes to compare radiant energies or photon fluxes at different wavelengths.

MEASUREMENT OF ABSOLUTE FLUXES

Ideally, absolute measurement of radiant energy can be carried out by calorimetric measurements under circumstances which guarantee that all photons in the beam are absorbed and that their energy is converted to thermal energy. This method is only practicable in the case of high-intensity sources in the visible part of the spectrum and is of very limited use in actual calibration procedures.

A second method is based on utilization of absolute sources or radiation standards. Two such sources are of importance. One is the black body, whose spectral distribution is uniquely specified for a given temperature in accordance with Planck's law. As a second source, there is the possibility of utilizing the emission from high-energy electrons accelerated in a betatron or synchrotron. As in the case of Planck's law, the spectral power radiated by a single electron is completely calculable; that is, the radiant power is expressible in absolute measure once the maximum electron energy and the orbital radius are known.

The use of the sources mentioned is beset with numerous experimental complexities. Electron accelerators are not generally available. So far as is known, the National Bureau of Standards' 180-Mev synchrotron is the only accelerator in the United States which is available for use as a far-ultraviolet source. There are many considerations which must be taken into account before

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the actual light beam may be adapted as a radiation standard: The effective number of radiators in the beam, the maximum energy of the electrons and the actual cross section of the electron beam must all be known. It is also necessary to see if the radial and vertical oscillations of the orbit are significant. The effects arising from the pulsed and polarized nature of the radiation must also be contemplated.

In the calibration of photographic emulsions or photoelectric devices of various kinds, one resorts to the use of thermocouples whose absolute response (microwatts per microvolt) is determined by first calibrating the thermoelectric detector against a radiation standard. This is done in the visible part of the spectrum and the calibration is assumed to hold in the far ultraviolet region also. In the case of strong radiations, thermocouple measurements have been made for wavelengths as short as 300A. Thermoelectric detectors do not possess 100% conversion efficiency, however, because diffuse scattering of the incident radiation and the kinetic energy carried away by the ejected photoelectrons are known to reduce the thermal conversion efficiency and give rise to errors which may be as high as 10%. Furthermore, when a thermocouple is used in the vacuum region, it becomes necessary to compare the response of the detector in the vacuum with its response obtained under atmospheric pressure. These conditions indicate that, even in the case of intense sources, the overall calibration of this type of detector is subject to large errors (approximately 20%) in the vacuum-ultraviolet region.

Photoionization detectors have recently become more popular. In this category we list gas-filled ion chambers with windows of lithium fluoride or similar material, ion chambers using differentially pumped entrance slits, and flow-type proportional or Geiger-Mueller (GM) counters equipped with special windows. Although the techniques used in photoionization detectors are in need of improvement, their development has reached the state where they could be considered as reliable detectors of photon fluxes.

For example, when the sensitive volume of a counter filled with a monatomic gas is illuminated by a beam of nearly monochromatic photons, incident photons with energies exceeding the ionization energy of the rarefied gas may interact with gas atoms to produce electron-ion pairs. (Ionization energies used are: He, 24.6 ev; Ne, 21.5 ev; Ar, 15.7 ev; Kr, 14.0 ev; and Xe, 12.1 ev.) For a sufficiently long path and high pressure, it may be assumed that the probability of stopping a photon in the counter is close to unity and that every photon that is absorbed will give rise to an electron-ion pair. In other words, the total cross section of the atom is equal to its photoionization cross section; thus, in the soft x-ray region, use of ionization chambers and Geiger-Mueller counters is based on the principle that photon interaction results only in photoionization. This is reasonable since for low photon energies, the Thomson and Compton scattering processes are negligible, pair production is impossible and second order effects such as double excitation are highly improbable.

In particular, if a Geiger-Mueller counter is to be used to measure absolute fluxes, one must be assured that each photoelectron will give rise to one (but not more than one) pulse. As a second working principle, it may be assumed that the probability of detecting the photoelectron is unity. The strongest support for this statement is based on the possibility of attaining a well
developed plateau when counting rate versus anode voltage is plotted. (One recalls that the output pulse is not dependent upon the charge liberated by the primary ionizing event.) Because of the long history of usefulness of Geiger-Mueller counters it is very probable that they also can adequately fill the need for photon-flux detectors for wavelengths shorter than 300Å. Above this wavelength Samson (Reference 1) has successfully applied ionization chamber measurements of absolute radiation intensities for wavelengths up to about 1000Å. The absence of convenient standard sources is no longer a serious handicap.

In the remaining portions of this paper, we describe the use of a Geiger-Mueller counter as applied to the problem of calibrating: (1) a specially designed photomultiplier and (2) certain photographic emulsions at wavelengths shorter than 300Å.

The specially developed photomultiplier tube is an open-window design. Photoelectrons from a tungsten cathode move along a cycloidal path in crossed electric and magnetic fields between two glass strips that are coated with a semiconducting, secondary-emitting oxide layer. One of the glass strips serves as a continuous dynode. Each photoelectron is multiplied to yield a pulse of approximately $10^6$ electrons at the anode.

DETERMINATION OF THE EFFICIENCY OF THE PHOTOMULTIPLIER IN THE 100Å TO 300Å RANGE

The purpose of this investigation was to attempt a measurement of the efficiency of the magnetic photomultiplier at wavelengths distributed over the soft x-ray region. The choice of wavelengths used was dictated by the availability of characteristic valence emission bands of beryllium, aluminum and chromium, roughly 100Å apart. These are quite steady, fairly intense, dc sources. They are therefore well suited for use with electronic detectors such as the Geiger-Mueller counter, that is, for studies which involve the measurement of photon fluxes. Recent techniques in the preparation of suitable plastic windows have led to successful use of the Geiger-Mueller counter as a device for determining absolute photon fluxes in wavelengths up to 300 angstroms. The performance and the reliability of this device are described elsewhere (Reference 2).

It suffices to say that for counting rates that are not excessive, the Geiger-Mueller counter operates very satisfactorily up to and beyond 300Å (its use at wavelengths lying between 20 and 100Å has already been demonstrated by Lukirskii and others (Reference 3)).

The measurements were performed in the following manner: The sources were operated at a pressure of $5 \times 10^{-6}$ Torr, a target current of 50 milliamperes and a target potential of about 1000 volts. A Geiger-Mueller counter and the photomultiplier were mounted within a grazing-incidence spectrometer. The detectors were attached to a rotatable table so that the Geiger-Mueller counter and the photomultiplier could be irradiated in succession by a narrow band of short wavelengths of continuous radiation produced in the target of the soft x-ray tube. Care was exercised to see that each detector was exposed to the same beam geometrically. The counting
rates of the detectors could then be compared. These rates are given by

\[ N_{GM} = N_0 \gamma_{GM} a_{GM} \]  

and

\[ N_{PM} = N_0 \gamma_{PM} a_{PM} \]  

where \( N_0 \) is the incident flux of photons and where \( N, \gamma \) and \( a \) denote, respectively, the counting rate, the efficiency, and the window area sensitive to photons. In Equations 1 and 2, the subscripts \( PM \) and \( GM \) refer to the Bendix photomultiplier and the Geiger-Mueller counter. From the relations expressed in Equations 1 and 2, it follows that the efficiency of the Bendix photomultiplier is given by

\[ \gamma_{PM} = \frac{N_{PM} a_{GM} \gamma_{GM}}{N_{GM} a_{PM}} \]  

Equation 3 expresses the desired efficiency at a specified wavelength since the quantities in its right-hand member are known.

Unavoidably, the characteristic valence band appears superimposed on the continuous radiation from the target. At a given wavelength, the radiation incident on the counter consists of the portion of the valence band, the bremsstrahlung at this wavelength, and contributions in higher orders from the continuum at shorter wavelengths. In these tests, the contribution from the continuum was subtracted and only the intensity due to the emission band was used. The emission bands involved were those of Be K (edge at 110Å), Al L\(_{2,3}\) (edge at 170Å) and Cr M\(_{2,3}\) (edge at 296Å). In each case, a trace of the entire band was recorded and counting rates were compared at wavelengths near band peaks or at corresponding wavelength positions. In this manner it was possible to avoid the effect of the contamination by C K\(_{\alpha}\) line in high orders, where such orders overlapped the emission band in question. Occasionally, it was convenient to deal with the second order spectrum of the emission band itself, because of the ease with which the band could be scanned.

Two points worth mentioning are that (a) the observed features of the bands were in good agreement with those determined by other experiments and (b) the angular separation between various emission bands as observed by the detectors was consistent with the calculated value of the angular separation.

In Table 1 the ratios of counting rates at the specified wavelengths are listed. The Geiger-Mueller counter was operated on its voltage plateau and the anode voltage applied

<table>
<thead>
<tr>
<th>Source</th>
<th>Wavelength (angstroms)</th>
<th>( N_{GM}/N_{PM} )</th>
<th>Independent Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe(^{55})</td>
<td>2</td>
<td>25 \pm 2.5</td>
<td>2</td>
</tr>
<tr>
<td>Be K</td>
<td>115</td>
<td>80 \pm 40</td>
<td>2</td>
</tr>
<tr>
<td>Al L</td>
<td>180</td>
<td>13.5 \pm 1.2</td>
<td>4</td>
</tr>
<tr>
<td>Cr M</td>
<td>305</td>
<td>4.0 \pm 1.0</td>
<td>5</td>
</tr>
</tbody>
</table>
to the photomultiplier was 1700 volts, except in the case of the Al L band where the anode voltage was 1650 volts. In each case, the areas of the entrance slits were determined by measuring the relevant dimension by a traveling microscope.

Before we can evaluate the photomultiplier efficiency, $\eta_{PM}$, we must determine the efficiency of the GM counter at the particular wavelength. The efficiency of the GM counter is given by

$$\eta_{GM} = \frac{1 - \exp(-\alpha(\lambda)p)}{K(\lambda)}$$

where $K(\lambda)$ is the attenuation due to the zapon window and the supporting mesh at the particular wavelength and $p$ is the gas pressure in torr. The expression in the brackets is the fraction of the incident number of photons/sec stopped by the counting gas (helium). This quantity is nearly equal to unity except at 115Å where the absorption cross section is quite low. The quantity $\alpha(\lambda)$ is related to the photoionization cross section $\sigma(\lambda)$ in accordance with

$$\alpha(\lambda) = 9.65 \ell \sigma(\lambda)/T$$

where $\ell$ is the absorption path length in cm, $\sigma(\lambda)$ is the cross section in megabarns (Mb) and $T$ is the absolute temperature of the gas in K. In formulating Equation 4 it has been assumed that photoelectrons from the back wall do not contribute appreciably to the observed count and that the discharge of the counter is due to photons being absorbed by the gas and giving rise to an ion pair. The validity of the last assumption is tested by the flatness of the voltage plateau of the Geiger-Mueller counter. One can correct for the effect of the added counts due to photoelectrons reaching the wall. This was done in dealing with $\eta_{PM}$ at a wavelength of 2Å.

The quantity $K(\lambda)$ was determined in the following way: while making the counter window, an identical zapon film was mounted on a frame. The attenuation of this film was measured at 115Å or at 180Å using the appropriate radiation. At 300Å, the attenuation was quite high and in addition the source was weak. In this case $K(\lambda)$ was computed by first obtaining the thickness of the film from the measured attenuation and published values (Reference 4) of the linear absorption coefficient of zapon. Having determined the film thickness by experiment at the short wavelength, one could then compute the attenuation at 305Å. The method was sufficiently precise for the present purposes.

The quantity $a(\lambda)$ defined by Equation 5 and appearing in Equation 4 is determined from the experimental values* (Reference 5) of $\sigma(\lambda)$ at the various wavelengths. In Table 2, the various quantities involved in evaluating the GM counter efficiency are summarized. With the information available in Tables 1 and 2, the efficiency of the photomultiplier is computed by use of Equation 3. The results are collected in Table 3 and are shown in Figure 1.

### Table 2
GM-Counter Efficiency.

<table>
<thead>
<tr>
<th>Wavelength $\lambda$ (angstroms)</th>
<th>Attenuation $K(\lambda)$</th>
<th>Cross-section $\sigma(\lambda)$ (megabarns)</th>
<th>$\alpha(\lambda)$ (torr$^{-1}$)</th>
<th>Gas Pressure $P$ (torr)</th>
<th>Efficiency $\eta_{GM}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>115</td>
<td>3.7</td>
<td>0.35</td>
<td>0.015</td>
<td>127</td>
<td>0.23</td>
</tr>
<tr>
<td>180</td>
<td>5.0</td>
<td>0.96</td>
<td>0.032</td>
<td>130</td>
<td>0.20</td>
</tr>
<tr>
<td>305</td>
<td>11.0</td>
<td>3.0</td>
<td>0.131</td>
<td>142</td>
<td>0.091</td>
</tr>
</tbody>
</table>

### Table 3
Photomultiplier Efficiency.

<table>
<thead>
<tr>
<th>Wavelength $\lambda$ (angstroms)</th>
<th>Efficiency $\eta_{PM}$ (percent)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2*</td>
<td>.23</td>
</tr>
<tr>
<td>115</td>
<td>.30 ± 0.1</td>
</tr>
<tr>
<td>180</td>
<td>1.7 ± 0.5</td>
</tr>
<tr>
<td>305</td>
<td>2.7 ± 0.5</td>
</tr>
</tbody>
</table>

*Efficiency at 2A was evaluated by taking into account the effect of photoelectrons from the back wall of the counter.

It appears that the photomultiplier is quite stable in its response and its characteristics have not changed in the course of several months. The efficiencies are plotted in Figure 1. (A comparison may be made with results obtained by Lukirskii (Reference 3) with a tungsten photocathode at 110A.) A second photomultiplier of the same type was tested at 180A and its efficiency was found to be about 4%. This represents a 2 to 1 variation. Variations of this order may originate in the response of the photocathode or in the collection efficiency of the secondary multiplication process.

### Calibration of Photographic Emulsions

In the spectral region of interest two types of special emulsions, Eastman SWR and Ilford Q, are commonly utilized. The purpose of these measurements was to correlate the photographic density with the number of photons/cm$^2$ required to produce this density. The correlation was carried out at various wavelengths using the emission bands of Be and Al and also the Lyman series lines of ionized helium. The ionized helium lines were excited in a Schuler lamp. Only the first four members of the series ($\lambda = 304A, 256A, 243A$ and 237A) were made use of in the calibration.
A piece of photographic film was mounted within the grazing incidence spectrometer close to the location of the Rowland circle. The film was inserted in the photon beam and exposed for a specified time. The photon flux was monitored before and after the exposure by the use of a Geiger-Mueller counter whose entrance slit could be made to occupy the same position as that occupied by the film. For purposes of photographic calibration, a series of different exposures was obtained at each discrete wavelength. In the case of the emission bands, the profile of the distribution was recorded photographically on film and also monitored by the Geiger-Mueller counter. The films were developed for 3 minutes in diluted (1:10) D19 developer at 68°F. The emulsion densities were obtained from microphotometer traces in the usual way and the corresponding photon densities were deduced from the counting rates and GM scan records.

The results obtained by the use of the two emission bands are presented in Table 4. The SWR emulsion shows a response which is essentially "flat" in the number of photons needed to produce a given density. The response of the Q emulsion is more nearly flat in energy.

In Figure 2 we display the various measurements of photon fluxes based on the He II lines. The solid curve is a relative photographic calibration determined independently by the use of spectral lines from a capillary spark discharge. This relative curve was moved horizontally to obtain the best fit to the points from the GM counter measurements, thus converting it from a relative to an absolute calibration.

Table 4

<table>
<thead>
<tr>
<th>Microphotometer Deflection d (scale divisions)</th>
<th>Photographic Density log (100/d)</th>
<th>Photon Flux (photons/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>(\lambda = 115\AA) &amp; (\lambda = 180\AA)</td>
</tr>
<tr>
<td>10</td>
<td>1.00</td>
<td>5.60 \times 10^6 &amp; 4.80 \times 10^9</td>
</tr>
<tr>
<td>20</td>
<td>0.70</td>
<td>3.35 &amp; 3.30</td>
</tr>
<tr>
<td>30</td>
<td>0.52</td>
<td>2.50 &amp; 2.40</td>
</tr>
<tr>
<td>40</td>
<td>0.40</td>
<td>2.00 &amp; 1.80</td>
</tr>
<tr>
<td>50</td>
<td>0.30</td>
<td>1.50 &amp; 1.40</td>
</tr>
<tr>
<td>60</td>
<td>0.22</td>
<td>1.20 &amp; 1.16</td>
</tr>
<tr>
<td>70</td>
<td>0.16</td>
<td>0.92 &amp; 0.92</td>
</tr>
<tr>
<td>80</td>
<td>0.097</td>
<td>0.69 &amp; 0.72</td>
</tr>
<tr>
<td>90</td>
<td>0.045</td>
<td>0.46 &amp; 0.50</td>
</tr>
</tbody>
</table>

(b) Ilford Q Emulsion S4125

<p>| | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>0.52</td>
<td>25 \times 10^9 &amp; 60 \times 10^9</td>
</tr>
<tr>
<td>40</td>
<td>0.40</td>
<td>4.00 &amp; 9.4</td>
</tr>
<tr>
<td>50</td>
<td>0.30</td>
<td>2.00 &amp; 4.5</td>
</tr>
<tr>
<td>60</td>
<td>0.22</td>
<td>1.20 &amp; 2.90</td>
</tr>
<tr>
<td>70</td>
<td>0.16</td>
<td>0.77 &amp; 2.00</td>
</tr>
<tr>
<td>80</td>
<td>0.097</td>
<td>0.50 &amp; 1.30</td>
</tr>
<tr>
<td>90</td>
<td>0.045</td>
<td>0.27 &amp; 0.68</td>
</tr>
</tbody>
</table>

Figure 2—Absolute calibration curve for Q1 emulsion S4125 at wavelengths of 304, 256, 243, and 237 angstroms.
REFERENCES


"The aeronautical and space activities of the United States shall be conducted so as to contribute ... to the expansion of human knowledge of phenomena in the atmosphere and space. The Administration shall provide for the widest practicable and appropriate dissemination of information concerning its activities and the results thereof."

—National Aeronautics and Space Act of 1958

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