

METHOD FOR ELIMINATION OF QUARTZ-FACE PHOTOTUBES
IN ČERENKOV COUNTERS BY USE OF WAVELENGTH-SHIFTER*

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Abstract

The feasibility of the use of organic wavelength-shifters in conjunction with normal glass phototubes and suitably transparent gases in Čerenkov counters is investigated. It is shown that such a combination gives performance at least the equal of that obtained from quartz-face phototubes.

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1. INTRODUCTION

The use of Čerenkov counters is widespread and essential in many areas of high-energy physics. Many such counters are used in the threshold mode and serve to identify particles at very high energies. In order to increase the number of effective photons from Čerenkov counters, experimenters have used quartz-windowed photomultiplier tubes whose response extends to almost 2000 Å. These quartz tubes are very expensive (about five times the cost of a normal tube at 5-inch diameter) and may have long procurement times. This paper presents a method for depositing and using organic wavelength-shifter materials which convert ultraviolet photons in the range 1100 to 3600 Å (for the recommended thicknesses) into emitted photons at wavelengths near 3850 Å, with very high quantum efficiency.

We also present experimental data on a test of these ideas in a Čerenkov counter using nitrogen gas at atmospheric pressure and an MgF_2 -overcoated, aluminized mirror in a test beam. Use of the wavelength shifter with a conventional phototube is shown to make the conventional tube at least the equal of a quartz-windowed tube in this counter. As expected, the 1- to 2-nsec decay of the shifter used did not noticeably affect the photomultiplier output pulse.

A proper wavelength shifter to be used in conjunction with the Čerenkov gas counter has to fulfill the following requirements:

- (a) Very high quantum yield of fluorescence for excitation wavelengths longer than λ_0 (λ_0 depends on the absorption properties of the gas used in the counter).
- (b) Fluorescence emission that matches the response of the normal S-11 cathode.
- (c) Short decay time of the excited state.

- (d) High transparency for its own fluorescence and for wavelengths longer than the fluorescence emission. This property will provide means to detect efficiently the Čerenkov radiation which is not absorbed by the shifter.
- (e) Stability against evaporation and aging, thus preventing deterioration of the converter and/or poisoning of the gas in the counter.

The most widely used of all the organic fluorescent converters--sodium salicylate--is not useful for the present purpose because of its relatively long lifetime, 8.5 to 10 nsec,¹ serious aging effects,² and low transparency for wavelengths longer than its own emission.

Several converters were investigated: Diphenylstilbene (DPS); p-Terphenyl (pTP); p-Quaterphenyl (QPh); 2,5 Diphenyloxazole (PPO); and P-Bis (2-(5-phenyloxazolyl)) benzene (POPOP).

2. EXPERIMENTAL METHODS

2.1. Sample Preparation

The organic scintillators were obtained from New England Nuclear, Pilot Chemicals Division, and used without any further purification. The measurements were performed on evaporated films, prepared by heating the various materials in an Mo evaporation boat covered by a thin wire mesh screen³ at a pressure of 10^{-5} mm Hg. A typical rate of deposition was 10^{-2} mg/cm²-min. The substrates were pyrex or quartz. The variations of the film thickness on the substrate were minimized by locating the substrate an appropriate distance above the evaporation boat. The thickness of a film was determined by weighing the substrate before and after the evaporation.

In order to prevent evaporation from the substrates of the scintillators, due to their high vapor pressure, the samples were coated with a protective thin layer

($\sim 200 \text{ \AA}$) of MgF_2 .⁴ This coating was done by evaporation immediately after the scintillator deposition, without venting the vacuum system. When the MgF_2 evaporation was performed at the rate of $\sim 100 \text{ \AA}/\text{min}$, the performance of the scintillator layer was not damaged by the coating. For lower rates of evaporation, an inferior performance was found in the vacuum UV region. The thickness of the MgF_2 layer was determined by a crystal thickness monitor technique.⁷

2.2. Lifetime Measurements

An oxygen-filled, nanosecond light source,⁸ combined with a 270-\AA Optics Technology interference filter, was used for excitation. The emission was detected by a 1P21 RCA phototube combined with a lucite lightguide. The decay curves were obtained on a Tektronix sampling oscilloscope. p-Terphenyl evidenced a decay time near 1 nsec, while diphenylstilbene showed a 2- to 3-nsec decay.

2.3. Emission Spectra

Emission spectra of some of the scintillators were measured on a Perkin Elmer MPF2A Fluorescence Spectrophotometer.

2.4. Wavelength Dependence of the Quantum Yield

The wavelength dependence of the quantum yields of the various scintillators was determined in the double beam attachment of a McPherson 225 Vacuum UV Monochromator. The readings of the photomultiplier viewing the scintillator were normalized to the readings of a photomultiplier viewing a sodium salicylate layer. Because sodium salicylate exhibits (to a good approximation) wavelength independent quantum yield,⁹ the normalization produced a result independent of the spectral distribution of the light source.

2.5. Measurement of the Response Curve of an Amperex 56 AVP Photomultiplier Coupled with a Wavelength Shifter

The wavelength shifter, on its substrate, was optically coupled¹⁰ by a thin

silicone grease layer to the front window of a 56 AVP photomultiplier, and viewed the exit slit of a Beckman DK-2A Ratio Recording Spectrophotometer. The anode current from the phototube was then recorded as a function of wavelength from 500 $m\mu$ to 250 $m\mu$ and normalized to the readings of a bare 56 UVP photomultiplier. For the wavelength region from 110 $m\mu$ to 250 $m\mu$, the wavelength dependence of the quantum yield obtained on the McPherson monochromator (see previous section) was utilized with a proper normalization.

2.6. Gas Absorption Measurements in the Vacuum UV Region

Absorption spectra of the various gases are available from the literature, but they are usually measured for very low pressures and for a very small optical path. Although it is difficult to deduce with accuracy from this data the detailed absorption edge at relatively high pressures (~ 1 atm) and long optical paths (1 m), those values which can be obtained are given in Table 1. We measured directly the complicated absorption spectrum of N_2 in our McPherson 225 Vacuum UV Monochromator. The measurement was performed in the following way: The H_2 light source was sealed with a LiF window and connected to the monochromator, which was filled with N_2 at 700 Torr. The light intensity transmitted through the 1-meter optical path of N_2 was recorded as a function of wavelength. This intensity was compared with the intensity of the light transmitted through the monochromator when it was under vacuum. For finding the absorption spectrum of N_2 at a pressure of 760 mm¹¹ the relationship $\frac{A_1}{A_2} = \frac{p_1}{p_2}$ (where A_i is the optical density for a pressure p_i of N_2) was used.

3. EXPERIMENTAL RESULTS

3.1. Optical Measurements

The relative quantum yields of the converters were determined. PPO and POPOP were found not to be suitable for the present purpose because of their

apparent lower quantum yield, accompanied, in the case of POPOP, by a high scattering for wavelengths longer than the fluorescence emission.

Lifetime measurements of samples of DPS, QPh, and pTP were performed. The shortest lifetime was that of pTP--between 1 and 2 nsec. The emission spectra of the above-mentioned scintillators are not noticeably attenuated by a pyrex window. The transmission of a 2.5-cm-thick pyrex window, along with the emission spectrum of a solid sample of pTP, is given in Fig. 1. This emission is considerably red-shifted in comparison to the solution emission given in Ref. 12. The emission spectra of DPS and QPh were not measured since the solution spectra,¹² which are usually blue-shifted with regard to the solid's emission, are not attenuated.¹³

No significant differences were found among the quantum efficiencies of pTP, DPS, and QPh; because of its lifetime, pTP seems to be the most suitable for the present purpose. The optimal thickness of the pTP layer is the minimum thickness required for an efficient fluorescence. Using thin layers has a double advantage: Scintillations excited directly by stray fast particles are negligible, while the optical transparency of the sample at wavelengths above the region of its absorption is improved. The recommended thickness of pTP layers is 0.2 - 0.3 mg/cm². At this thickness, less than 6% of the light in longer wavelengths than the emission is lost. For thicknesses less than 0.2 mg/cm², a decrease in emission for excitation wavelengths in the 2200- to 3400-A region was found.

In order to compare the relative efficiencies of the 56 AVP photomultiplier coupled with a wavelength shifter (this combination will be called 56 AVP' for sake of convenience) and a 56 DUVP photomultiplier, the response curve of the tube plus scintillator was constructed in a fashion explained in detail in Section 2.5. (The response curve was calculated for a typical thickness

of 0.5 mg pTP/cm².) Defining the response of the combination as $R_{AVP'}(\lambda)$, the number of photons of Čerenkov radiation per unit wavelength interval as $N_C(\lambda)$, the transmission of gas x as T_x , the integral

$$\int R_{AVP'}(\lambda) N_C(\lambda) T_x(\lambda) d\lambda \equiv I_1$$

was calculated. Defining the response of the 56 DUVP photomultiplier as $R_{DU}(\lambda)$, the integral

$$I_2 \equiv \int R_{DU}(\lambda) N_C(\lambda) T_x(\lambda) d\lambda$$

was calculated. The relative values of $R_{DU}(\lambda)$ were taken from Ref. 14, while at an arbitrary wavelength λ' --in our case, $\lambda' = 450 \text{ m}\mu$ --(this wavelength has to be in the visible), the equality $R_{DU}(\lambda') = R_{AVP'}(\lambda')$ was set. The physical meaning of this equation is normalization of the output of the two tubes, when viewing the same visible light source, which, in our case, was a nanosecond-pulsed lamp.

The values of I_1 and I_2 were calculated for various gases, and they are given in Table 1. Except for the absorption data of N_2 , which were measured as described in the experimental section, the other absorption data are deduced from the information available in the literature. Because of the existing inaccuracy in some of the available data, the absorption spectra of the more interesting gases will be measured directly and published separately. It is worth mentioning that for a counter using mirrors, the wavelength-dependent reflection function must be included in the calculations. A successful experiment to compare the relative efficiencies of 56 AVP' and 56 DUVP in a Čerenkov counter was performed.

3.2. Čerenkov Counter Beam Tests

The wavelength shifters were experimentally tested in a Čerenkov counter

shown schematically in Fig. 2. In order to simplify construction, the counter was run at atmospheric pressure. Nitrogen gas was used as a radiator because it has excellent ultraviolet transmission properties and because it can easily be obtained at very high purity. A constant flow of nitrogen gas was supplied to the counter by evaporation from a liquid nitrogen dewar. The oxygen contamination was monitored in the effluent gas from the counter by means of a hot zirconium-oxide detector. The oxygen impurity was typically 12 ppm at a flow rate of 4 scf per hour.

The particle beam through the Čerenkov counter was defined by 3 scintillation counters and 2 iron collimators to a diameter of about 1 cm. The Čerenkov light, emitted by high-energy electrons (3 - 15 GeV) over a length of 2 meters, was reflected by a front-surface spherical mirror (radius = 122 cm) and focussed at the entrance window of the photomultiplier. The mirror was fabricated by slump-molding plexiglass. It was coated with 1000 Å of aluminum deposited at a rate of about 30 Å per second, followed immediately by 250 Å of MgF₂ deposited at a rate of about 8 Å per second.

The 3 scintillation counters were used to trigger a linear gate which passed the Čerenkov counter pulse to the input of a 128-channel analyzer. The pulse analyzer spectrum was then displayed on the oscilloscope and photographed for permanent record.

In this experiment, 2 unselected Amperex phototubes were used: a 56 AVP and a 56 DUVP. The gains of the tubes were equalized at about 4500 Å by using a pulsed nsec light source (PEK 118) filtered by a Kodak Wratten neutral-density filter of 10× attenuation located at the beam entrance end of the Čerenkov counter.

Two different wavelength shifter compounds were evaluated: p-Terphenyl and Diphenylstilbene. Each of the shifters was evaporated onto a 1/8-inch thick

Vycor disc and immediately overcoated with about 250 Å of magnesium fluoride. During the experiment, the appropriate disc was then optically coupled to the window of the phototube with silicone grease. The thickness of the pTP layer was 0.23 mg/cm² and the thickness of the DPS was 0.08 mg/cm². The results for the two wavelength shifter compounds were quite comparable; however, a 20% further increase in average output was obtained by coating the 56 AVP directly with 0.2 mg/cm² of pTP and 270 Å of MgF₂. This result is shown as a curve in Fig. 3. Also shown in Fig. 3 are the responses of the 56 DUVP and the uncoated 56 AVP. It is worth mentioning that in spite of the efficient scintillation properties of nitrogen, the amount of scintillation light was negligible in comparison to Čerenkov light. This fact was clearly demonstrated when the pulse height spectrum of 3- to 6-GeV π mesons was compared with that of the 3- to 15-GeV electrons. The intensity of the Čerenkov light from electrons is at least 5 times greater than the intensity of the scintillations from π mesons. In Fig. 3, the vertical scale is arbitrary and only the shapes of the spectra are meaningful. The average of the pulse height spectrum from the 56 AVP was increased by a factor of 3 by coating the tube with wavelength shifter. The larger width of the 56 AVP distribution (compared to the 56 DUVP) is due mainly to the lower quantum efficiency of the 56 AVP and not due to additional broadening introduced by the wavelength shifter, which has a quantum efficiency near 1. (The broader spectrum also manifests itself in the pulse height distribution from the calibrating light source.) It is easily seen from Fig. 3 that coating a 56 AVP tube makes it slightly superior in Čerenkov light converting efficiency to the much more expensive 56 DUVP.

With pTP, no change in the decay time of the output pulse from the phototube could be seen, while with DPS, a slight increase in the pulse decay time could be

detected. A study of aging effects on the shifters in the presence of various gases, which might be potential Čerenkov gas counter fillers, is in progress.

4. CONCLUSIONS

We have demonstrated that normal glass phototubes, when used with properly deposited organic wavelength shifters and suitably transparent counter gases, give performances equal or superior to quartz-face phototubes in Čerenkov counter applications. Other investigators²¹ have reported improved efficiencies with the longer decay time material, QPH.

References

1. G. K. Herb and W. J. Van Sciver, Rev. Sci. Instr. 36, 1650 (1965).
2. J. A. R. Samson, J. Opt. Soc. Am. 54, 6 (1964).
3. The mesh screen was found to be very useful in preventing explosive scattering of the material from the evaporation boat.
4. This thickness of MgF_2 is not expected to absorb the incident radiation.⁵ MgF_2 was preferred to LiF because of the hygroscopic property of the latter.⁶
5. J. A. R. Samson, Techniques of Vacuum Ultra Violet Spectroscopy, John Wiley and Sons, Inc., 1967, p. 183.
6. D. A. Patterson and W. H. Vaughan, J. Opt. Soc. Am. 55, 1078 (1965).
7. A. H. F. Muggleton and F. A. Howe, Nucl. Instr. and Methods 28, 242 (1964).
8. L. Hundley, T. Coburn, E. Garwin, and L. Stryer, Rev. Sci. Instr. 38, 488 (1967).
9. K. Watanabe and E. C. Y. Inn, J. Opt. Soc. Am. 43, 32 (1953).
10. J. B. Birks, The Theory and Practice of Scintillation Counting, MacMillan Company, New York, 1964, p. 98.
11. The monochromator was not filled to 760 Torr N_2 , because slight fluctuations in that pressure would cause O_2 contamination.
12. I. B. Berlman, Handbook of Fluorescence Spectra Aromatic Molecules, Academic Press, 1965.
13. This property is very important for highly pressurized Čerenkov counters, which can use pyrex windows coated on the high-pressure side by a wavelength shifter.
14. Amperex Electron Tubes Data Handbook, 1971.

15. J. V. Jelley, Čerenkov Radiation and Its Applications, Pergamon Press (1958), p. 239.
16. V. P. Zrelov, Čerenkov Radiation in High Energy Physics, Vol. II, translated from the Russian, Israel Program for Scientific Translations, Jerusalem, 1970.
17. H. Okabe and D. A. Becker, J. Chem. Phys. 39, 2549 (1963).
18. Ref. 5, p. 131.
19. T. Liu, G. Moe, and A. B. F. Duncan, J. Chem. Phys. 19, 71 (1951).
20. E. L. Garwin and A. Roder, Nucl. Instr. and Meth. 93, 593 (1971).
21. A. M. Blagorodov, V. V. Vladimirkii, G. K. Kukavadze, B. Ya. Kuznetsov, and N. V. Novikova, Pribory i Tekhnika Éksperimenta 4, 59, (1968).

Table 1

Gas	$\eta = n - 1$	Absorption edge (\AA)	I_1/I_2
Methane	$4.41 \times 10^{-4(15)}$	1640 ^(a)	1.3
Propane	$1.005 \times 10^{-5(15)}$	1850 ^(a)	1.06
Butane	$14.37 \times 10^{-4(d)}$	1960 ^(a)	0.95
Krypton	$4.27 \times 10^{-4(15)}$	1236 ^(b)	1.8
Xenon	$7.02 \times 10^{-4(15)}$	1470 ^(b)	1.5
SF ₆	$7.85 \times 10^{-4(16)}$	1473 ^(c)	1.5
Freon 12	$11.52 \times 10^{-4(d)}$	2220 ^(e)	0.7
Freon 13	$7.99 \times 10^{-4(d)}$	1970 ^(e)	0.9
Freon 13B1	$10.07 \times 10^{-4(d)}$	2600 ^(e)	0.68
CO ₂	$4.5 \times 10^{-4(15)}$	1920 ^(e)	0.97
N ₂	$2.97 \times 10^{-4(15)}$	1400 ^(e)	1.9

Caption for Table 1

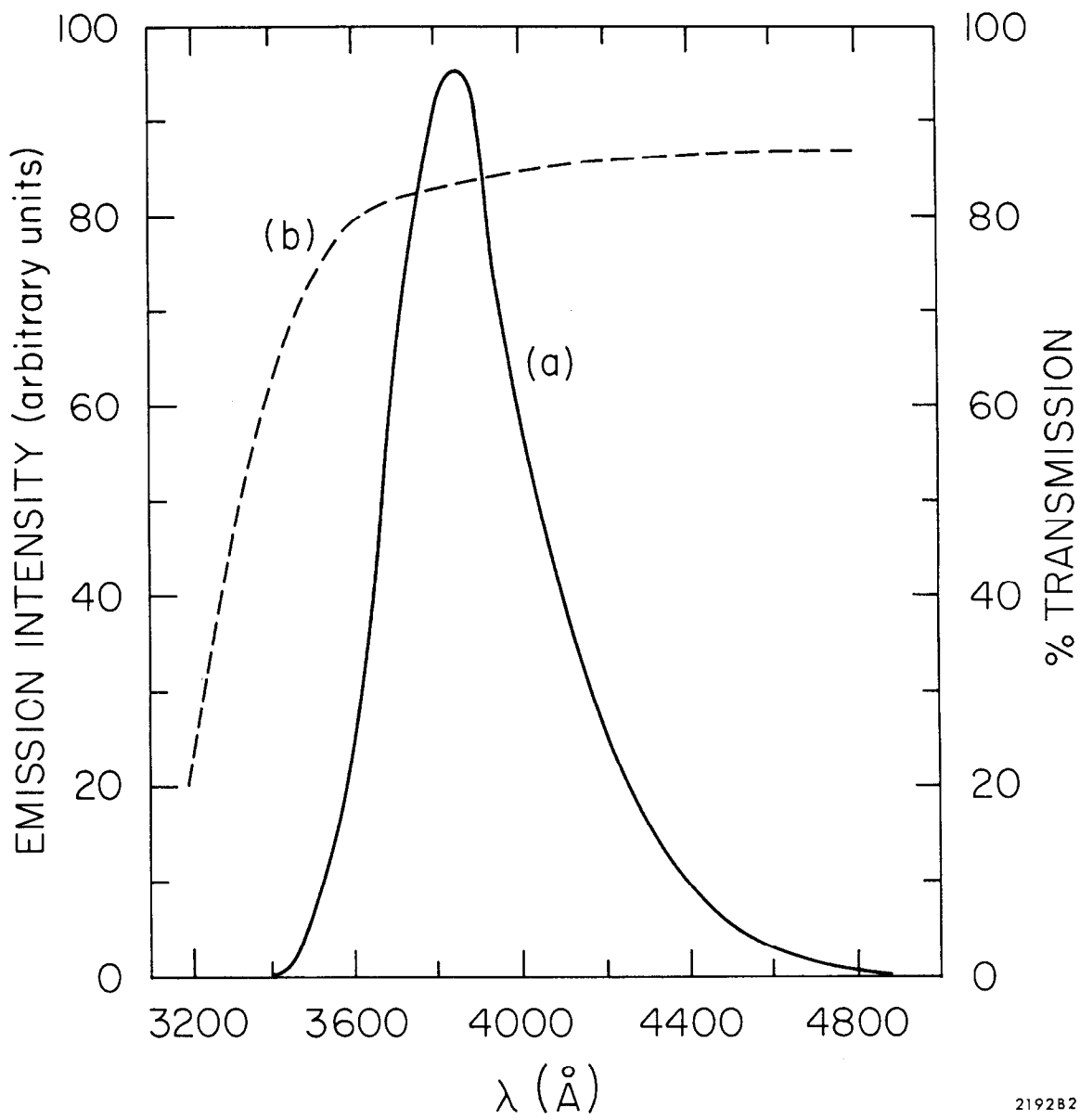
The calculated photocathode current, I_1 , resulting from Čerenkov light incident on a 56 AVP photomultiplier coated with a pTP wavelength-shifter divided by the calculated photocathode current, I_2 , resulting from the same Čerenkov light incident on a 56 DUVP photomultiplier, is given for different gases. The indices of refraction of these gases, measured at a wavelength $\lambda = 5893 \text{ \AA}$ at 0° C and 760 mm Hg, are listed, together with the extrapolated absorption edges for those gases for which accurate absorption data were not available.

- (a) The absorption edge of methane is calculated to be at 1640 \AA , based on the following: According to Ref. (17), $k = 1 \text{ atm}^{-1} \text{ cm}^{-1}$ at 1440 \AA where k is defined by the relationship $I = I_0 e^{-kpx}$. Define the absorption edge for a given pressure p and a given optical path x as the wavelength where the equality $kpx = 1$ exists. Extrapolating from the known rate of decrease of k with increasing wavelength up to 1440 \AA toward longer wavelength regions, $k = 10^{-2} \text{ atm}^{-1} \text{ cm}^{-1}$ (absorption edge for 1 atm and optical path of 1 meter) at 1640 \AA . Propane and Butane are calculated similarly from (17).
- (b) The absorption edges of Krypton and Xenon are assumed to be at the positions of the raie ultime (the first resonance line) at 1236 \AA^{18} and 1470 \AA^{18} , respectively.
- (c) The absorption edge of SF_6 is assumed to be at 1473 \AA . This assumption is based on the following: The maximum of the first electronic transition is found by (19) at 1054 \AA with a width of 9000 cm^{-1} . Define the absorption edge for this particular case as the position shifted from the maximum 3 times the width towards longer wavelengths.
- (d) The indices of refraction have been calculated by the Lorenz-Lorentz law using atomic refraction values from the Handbook of Chemistry and Physics, 45th edition, 1965.

(e) The absorption edges, defined as in (a), are given only for sake of comparison. In order to obtain the values of I_1/I_2 , the real absorption spectra were folded into the Čerenkov light spectrum. The absorption spectra of Freon 12, Freon 13, and CO_2 are taken from Ref. (20); the absorption spectrum of Freon 13B1 was measured by the authors of Ref. (20). The absorption of N_2 was measured by us.

Figure Captions

- Fig. 1. Curve "a" is the emission spectrum of a polycrystalline sample of p-Terphenyl. The wavelength of the exciting radiation is 3100 Å. Curve "b" represents the transmission of 2.5-cm-thick pyrex window.
- Fig. 2. A schematic of the Čerenkov counter.
- Fig. 3. Pulse height spectra. Curve "a"—56 AVP coated directly with 0.2 mg/cm² p-Terphenyl and 270 Å MgF₂. Curve "b"—56 AVP without coating. Curve "c"—56 DUVP. The vertical scale is in arbitrary units.



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Fig. 1

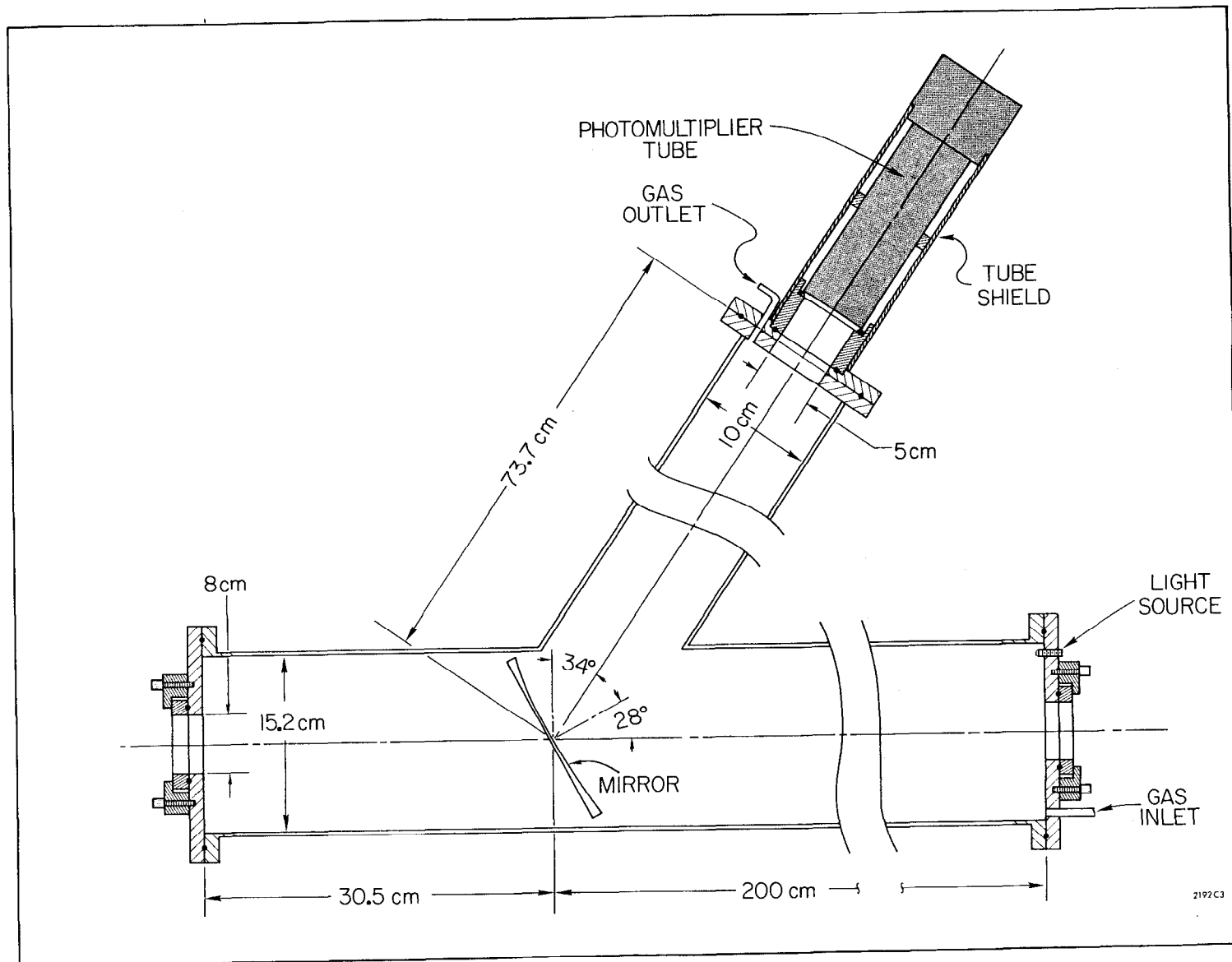


Fig. 2

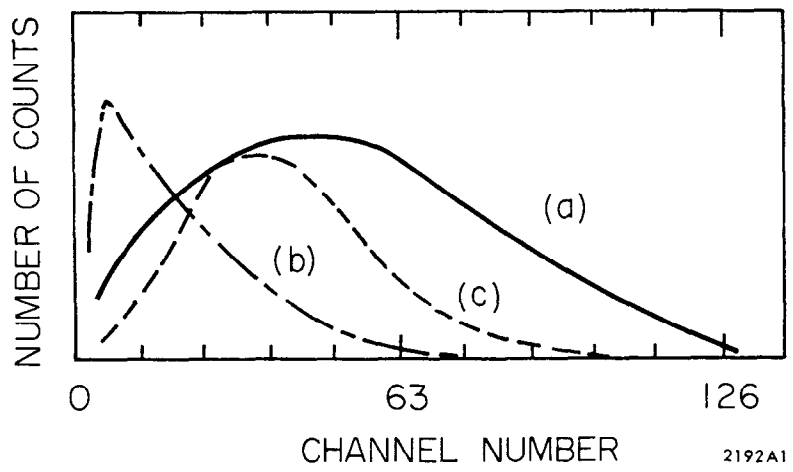


Fig. 3

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