COUPLING OF A BaF₂ SCINTILLATOR TO A TMAE PHOTOCATHODE
AND A LOW-PRESSURE WIRE CHAMBER

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with Appendix by G. Kneller⁴

ABSTRACT

The short wavelength component of a BaF₂ scintillator has been successfully coupled to a tetrakis(dimethylamine)ethylene (TMAE) photocathode and a low-pressure wire chamber. An energy resolution of 28.5% FWHM has been measured for protons losing about 18 MeV in the crystal. A timing resolution of 540 ps FWHM has been measured for 350 MeV α -particles. We foresee a new generation of calorimeters with good spatial, temporal, and energy resolution, able to work at high rates. Other possible applications are also discussed.

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1. INTRODUCTION

The modern work in the field of UV light detection with a wire chamber began with the suggestion by Séguinot and Ypsilantis [1] that an admixture gas with a low photo-ionization potential I_g might be added to a wire chamber with an optical window. This was then demonstrated by Charpak et al. [2] with the use of triethylamine (TEA) in a multistep chamber, imaging Cherenkov photons. The threshold of TEA is 7.5 eV (165 nm), which requires the use of CaF_2 windows. The next advancement was made by Anderson [3] with the introduction of tetrakis(dimethylamine)-ethylene (TMAE), with a threshold of 5.4 eV (231 nm), which allowed the use of quartz windows. The primary interest in photosensitive wire chambers has been in the detection of the light from xenon-filled gas scintillation proportional counters and Cherenkov ring imaging. Although TMAE is somewhat hampered by its low vapour pressure (0.35 Torr at 20 °C) [4], no gas with a lower value of I_g has yet been found.

In the search for gases with low I_g , it was noted that the photocurrent threshold E_{th} of the material in solution is much lower than its value of I_g . A value of $E_{th} = 3.54$ eV (350 nm) has been measured for TMAE dissolved in tetramethylsilane [5]. This led to the demonstration of a liquid TMAE photocathode coupled to a low-pressure wire chamber, by Anderson [6] who also detected the UV photons from BaF_2 . It should also be noted that the same author earlier observed the detection of the light from BaF_2 with a single-wire proportional counter filled with TMAE, and a mixture of argon (90%) and methane (10%) [4]. It was believed that the light was detected by the TMAE in the gas phase, but, as will be shown by this work, this was in fact the first demonstration of a TMAE photocathode. The poor efficiency obtained in that early work was probably due to the use of a counter working at atmospheric pressure.

The reasons for the interest in coupling a high-Z scintillator to a wire chamber are many. It would, in principle, allow the detection of high-energy particles or photons (e.g. calorimetry, or positron emission tomography) without

the restraints imposed by photomultiplier tubes. One could also envisage systems that are very compact, offering good timing and spatial resolution.

2. TEST DETECTOR

Figures 1a and 1b show a schematic of the test detector and a detail of the electrode structure. The detector consists of an aluminium chamber sealed with Viton O-rings. The scintillator is a BaF_2 crystal, 130 mm in diameter and 26 mm thick, in one of two configurations: i) the first surface is aluminized and the second surface is in contact with a 90% transparent mesh; or ii) the first surface is clean and the second surface is coated with a thin surface of NiCr as a transparent conductive surface. After an 8 mm drift space, there is an amplification region consisting of two 90% transparent meshes and an anode made of 20 μm wires with a 1 mm pitch. The cathode-to-anode spacing is 3 mm. After a second drift space of 4 mm, there is a temperature-controlled aluminium surface. The surface of the BaF_2 and that of aluminium are kept at ground potential, and the cathode meshes are connected to a common power supply. The vacuum and gas systems are connected to one port, and a temperature-controlled flask of TMAE liquid is accessed through a second one.

In the original concept of the test detector, the photosensitive surface would be a condensed layer of TMAE on the aluminium surface. The collection and amplification regions were separated to give greater flexibility in the operation parameters. In most cases the gas filling was 3 to 9 Torr of pure isobutane. Methylal was also used as a counter gas.

The test detector has a small, thin, aluminium window to allow the entry of low-energy γ -rays. Their interaction with the BaF_2 was used to provide single photoelectrons from the cathode.

3. ADVANTAGES OF LOW-PRESSURE COUNTERS

There are several reasons that have led us to use low-pressure counters (see refs. 7 and 8 on the subject). The first reason is that high-pressure gas has been found to decrease the collection efficiency of photoelectrons from a

photocathode. As discussed in the Appendix to this paper, the use of low-pressure organic gases gives the highest quantum efficiency from a photocathode.

A second advantage of low-pressure counters is the good timing characteristic. The dependence of time resolution with counter gas pressure is shown by the work of Breskin, Tserruya and Zwang [7] (fig. 2). As will be discussed in the next section, the component of BaF_2 to which TMAE is sensitive has only a 600 ps decay time. Thus a low-pressure counter seems to be a natural choice if the good timing characteristics of BaF_2 are to be maintained.

Another advantage of low-pressure counters is that they work with pure "quench" gases such as isobutane or methylal. Other gases such as argon/methane mixtures, or even pure methane, produce UV photons in the gas amplification process. These photons are to be avoided since high gain is desired. With isobutane we were able to achieve gains of the order of 10^6 without photon feedback.

Low-pressure counters have very low sensitivity to minimum ionizing particles, which will lose, on the average, one electron per centimetre in 3 Torr of isobutane. Thus the photon detector is almost exclusively sensitive to the UV photons and not to the particles in the shower.

Finally, low-pressure counters have very high ion drift velocities. The ion occupation time is low and, since the photons are detected over some area, the ion density is also low. Thus BaF_2 coupled to a low-pressure counter should be able to work at very high rates.

4. BaF_2 AS A SCINTILLATING CRYSTAL

The scintillation properties of BaF_2 have recently gained in interest with the discovery of the very short decay constant of one of its two emission components [9]. The first scintillation component to be discovered had an emission peak at 310 nm and a decay constant of 620 ns. Later, a fast component containing about one quarter of the light was discovered, with an emission peak at 225 nm and a decay constant of only 0.6 ns. Using this fast component, Laval et al. [9] have obtained a timing resolution of 112 ps FWHM for ^{60}Co photons. They have

also measured the emission spectra of BaF_2 that are shown in fig. 3. The transmission of 27 mm of BaF_2 is also included. From an early measurement done with filters, the detection threshold for a TMAE photocathode was estimated to be about 4.3 eV (290 nm). Thus it is primarily sensitive to the fast component.

Before the demonstration of the detection of the light from BaF_2 with a wire chamber, BGO and NaI(Tl) were the only high-Z crystal scintillators considered for calorimetry. Some properties of BaF_2 , BGO, and NaI(Tl) are listed in table 1 (refs. 9-12), and it can be seen that BaF_2 has properties that fall somewhere between the other two, with the notable exceptions of decay constant and index of refraction. Its short decay component is 400 to 500 times faster, whilst its smaller index of refraction allows a larger fraction of the light to leave the crystal. The light yield of the fast component is about the same as that of BGO but is considerably less than NaI(Tl) . In general, BaF_2 is a strong, non-hygroscopic crystal, with the greatest resistance to high-energy radiation of all the fluorides listed in the Harshaw Optical Crystal Catalog. It also has the hardest UV scintillation light of any crystal known to us.

5. PERFORMANCE

In this section we will discuss data taken with a parasitic test beam at Saclay. The BaF_2 crystal with the aluminized first surface was used along with the ground plane of 90% transparent mesh. The aluminium surface was maintained at 2 °C, with a thin layer of TMAE liquid condensed on it. As will be shown in the next section, this is not the best configuration. The gas filling was 3 Torr of isobutane.

The first tests were made using α -particles with a maximum energy of 350 MeV. Figures 4 and 5 show the nature of the pulses from the test detector. The first figure, with a time scale of 4 ns/division, shows the electron component of the signal. The rise-time is about 10 ns. Figure 5, with a time scale of 0.5 μs per division, shows an ion drift time of less than 1.5 μs .

Again using α -particles, the timing resolution between the test detector and a 1 cm plastic scintillator was measured. The timing spectrum is shown in fig. 6. The time difference between the two peaks is 2 ns, and the resolution is only 540 ps FWHM.

For an energy resolution determination, 940 MeV protons were used, depositing about 18 MeV in the BaF_2 crystal. Figure 7 shows a pulse-height spectrum taken at an instantaneous rate of about 2×10^5 counts per second. The energy resolution was measured to be 28.5% FWHM. Assuming an $E^{1/2}$ dependence on energy resolution, this implies a resolution of 3.8% FWHM (or $\sigma = 1.6\%$) at 1 GeV loss in the detector. Thus at high energies the energy resolution of a BaF_2 calorimeter will probably not be limited by photon statistics but by physical factors.

An estimate was also made of the number of photoelectrons detected per MeV. This was done by comparing the pulse height of cosmic rays depositing about 15 MeV in the BaF_2 , with a single photoelectron spectrum generated with 59.5 keV γ -rays from ^{241}Am . The best estimate for the configuration discussed here was 11.3 ($\pm 20\%$) photoelectrons per MeV or 11.3×10^3 photoelectrons per GeV. Assuming 2×10^3 photons per MeV produced in the crystal, a critical angle of 40° , and a 30% enhancement in light collected by the aluminized surface, we estimate the quantum efficiency of the condensed TMAE photocathode to be about 4%.

6. TMAE AS A PHOTOCATHODE

After the measurements described in the previous section, it was discovered that it was not necessary to condense TMAE on a cold surface. If TMAE gas is introduced into an evacuated chamber, all metal surfaces become photosensitive within seconds. Depending on the surface and the counter temperature, it may take from ten minutes to tens of hours to remove this photosensitivity by evacuation.

Upon realizing this fact, we replaced our BaF_2 crystal with the second one coated with a thin layer (15–25 Å) of NiCr. This was to provide a UV transparent, conductive surface for the TMAE. For the three figures that follow (figs. 8, 9, and 10), with time scales of 50 ns/division, the evacuated detector was kept at

room temperature and exposed to the temperature-controlled source of TMAE liquid. The counter was then filled with 9 Torr of isobutane. Figure 8 shows cosmic-ray events with the TMAE at 2 °C. The three bumps show the arrival of the photoelectrons from i) the electrical cathode mesh; ii) the aluminium surface, and iii) the BaF₂ surface (see fig. 1b). Figure 9 shows the cosmic-ray events with the TMAE at 20 °C. Finally, fig. 10 shows the response of the detector after 12 hours of evacuation. The BaF₂ crystal is still sensitive. After another 24 hours of evacuation, this effect disappeared. It should be noted that the vertical scale on these three figures has no absolute significance, since the gain of the detector is dependent on the amount of TMAE present; thus the scale was adjusted to give events of similar heights. Although these figures are representative, there was considerable variation in the ratio of the bumps seen from event to event.

On first looking at fig. 8, one notices that the contribution of the 90% transparent mesh is disproportionately high. Some of this may be because of enhanced emission due to the stronger electric field at the surface of the mesh [13]. But we also have evidence that suggests that the electrons coming from this mesh have slightly higher gain than those from the other surfaces. If this is the case, such cathodes should be avoided since they would degrade the energy resolution. Also, if it is not enhanced emission, such cathodes would seem to be detrimental where optimum timing is desired.

We now see that the use of TMAE as a photocathode is not as difficult as was first anticipated. There are no detector cooling requirements. Since the amount of TMAE absorbed on the surfaces seems to be dependent only on the pressure of the TMAE, flow counters seem to be feasible with the counter gas simply bubbled through TMAE at a controlled temperature. Also, since the TMAE can be deposited directly on the BaF₂ surface, good position resolution should be possible for imaging detectors using these crystals.

Unfortunately we have not determined the number of photoelectrons detected per MeV of energy loss in the BaF₂ in this new configuration. One would hope that with the NiCr and TMAE directly on the BaF₂, less light will be trapped in

the crystal and thus more will be detected. We have also not been able to evaluate the dependence of quantum efficiency of the photocathode on the electric field strength. There is still much work to be done in understanding TMAE as a photocathode.

7. COST OF BaF₂

It is difficult to discuss costs in a publication, but when one talks of calorimetry the quantity is in tons, and costs are important (1 ton of BaF₂ is a cube with a 60 cm side). At present we pay \$6 per cubic centimetre for cut and polished BaF₂ crystals, 120 mm in diameter by 25 mm thick. This is about 25% of the price for large crystals of BGO. But the potential for reduction in price is dramatic. The price for pure BaF₂ powder (of the order of 1 ton from Fluka, Switzerland) is \$0.018 per cubic centimetre of crystal. The Ge in BGO costs \$1.42 per cubic centimetre (in the form of GeO₂ from China).

It is also much easier to produce BaF₂ crystals than BGO. Six years ago, about 40 kg of BaF₂ were produced at CERN. The production and material costs were low, and we have found that this BaF₂ is within a factor of 2 of the light output of the best BaF₂ available. There is hope that for large instruments the cost of the BaF₂ will be greatly reduced over the present price for small orders.

8. ADVANTAGES OF A BaF₂ PLUS TMAE CALORIMETER

One of the greatest advantages of BaF₂ plus TMAE as a calorimeter is that BaF₂ can be subdivided into slices of 1 or 2 radiation lengths, separated by 4 to 6 mm of low-pressure counter. This will allow the longitudinal shower development to be observed in a compact instrument with scintillator energy resolution. This would also allow lower quality material to be used, since self-absorption will be much less of a problem in 2 radiation lengths pieces than in one 22 radiation lengths piece. The insertion of thinner pieces of BaF₂ coupled to imaging detectors would also allow the lateral development of the shower to be monitored and would give some two-particle identification capability.

The speed of a BaF₂ plus TMAE calorimeter will also be a great advantage. We have demonstrated good energy resolution at a rate of 2×10^5 counts per second. With the proper electronics, this number could be substantially increased. Also, with the large energies deposited in a calorimeter, and with several BaF₂ crystals available for timing, the 0.5 ns timing resolution reported here should easily be attainable in a practical instrument.

The performance of this instrument in a magnetic field has not, as yet, been demonstrated. It will certainly be less sensitive than photomultipliers, and may possibly work in very high magnetic fields with only little effect.

There is also reason to hope that the price of large quantities of BaF₂ will be substantially reduced. This would allow the construction of instruments, at reasonable cost, that can handle very high rates whilst giving more information than other calorimeter designs.

9. OTHER APPLICATIONS

If the light detection efficiency can be improved for the BaF₂ plus TMAE system, many applications suggest themselves. Using thin BaF₂ crystals and an imaging proportional counter, imaging γ -ray detectors should be possible for γ -ray astronomy and nuclear medicine. One can also foresee applications in positron emission tomography. There are also possible applications in radiation detection and dosimetry, where the gaseous detector can be used either in the ionization mode or with gain.

We also see an application for TMAE photocathodes in Cherenkov ring imaging. Although the quantum efficiency is lower than for TMAE gas, the lower threshold allows a greater variety of radiators to be used. One possible configuration for the use of a TMAE photocathode for Cherenkov ring imaging is to have planes of wires, with collection of the photoelectrons on a single plane of anodes. This would allow the imaging of a greater number of points in an event, since electrons from different planes will arrive at different times. If we are seeing enhanced emission from wires, the quantum efficiency may be much higher than the 4% quoted above.

10. CONCLUSIONS

We have presented here a technique that offers new prospects in electromagnetic calorimetry. BaF_2 coupled to a TMAE photocathode has the potential of yielding calorimeters capable of high rates, energy resolution limited only by leakage, subnanosecond timing, and ability to display both longitudinal and lateral shower development. The work presented here also has applications in many other fields, such as nuclear medicine, γ -ray astronomy, and Cherenkov ring imaging. But there is a great deal more work to be done. We need to find a source of large pieces of BaF_2 at a moderate price. The effects of magnetic fields, collection voltage, and surface materials need to be studied. A search for photosensitive materials other than TMAE must also be pursued. But it seems clear that the use of condensed gases as a photosensitive surface has a promising future.

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Table 1

Properties of three scintillators*)

	BaF ₂	BGO	NaI(Tl)
Density (g/cm ³)	4.9	7.1	3.7
Radiation length (cm)	2.1	1.1	2.6
dE/dx (min.) (MeV/cm)	~ 6	8	4.8
Linear attenuation coefficient at 511 keV (cm ⁻¹)	0.47	0.92	0.34
Peak emission (nm)	225 310	480	410
Decay constant (ns)	0.6 620	300	250
Index of refraction	1.56	2.15	1.85
Light yield (photons/MeV)	2×10^3 6.5×10^3	2.8×10^3	4×10^4
Hygroscopic	No	No	No

*) See refs. 9-12.

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APPENDIX

EXTRACTION OF PHOTOELECTRONS INTO A GAS

(by G. Kneller)

For the work presented above, it was considered necessary to have at least a qualitative understanding of the effect of gas pressure and composition on the collection of photoelectrons from a photocathode. To study this, we constructed a photodiode consisting of a CaF_2 window followed by a grounded mesh and an aluminium photocathode. The cathode-to-mesh distance was 5 mm. The photocathode was held at a negative potential and the mesh at ground. A UV light source with an intensity that varies at 100 Hz was used. The output voltage was measured at the photocathode with an a.c. coupled amplifier. The photodiode was connected to a vacuum and gas-filling system.

The gases evaluated were helium, argon, methane, and isobutane. For all tests the instrument was first evacuated for several hours, and the output voltage (peak-to-peak) was measured as a function of collection voltage. All tests made with gases were then normalized to the vacuum measurement at the same collection voltage. An efficiency of the same value as the vacuum measurement would have a value of 100%.

With the exception of helium, we found that after an exposure to a gas, the vacuum measurement was not reproducible. This effect varied with gas type, pressure, and length of filling time. The effect was that of greater electron yield after exposure to a gas.

We present here two sets of measurements that are the most reproducible and show the effect of gas pressure and composition.

Figure A.1 shows the effect of helium for various pressures on the collection of photoelectrons. Again, all measurements have been normalized to the vacuum measurements. As can be seen, even 3 Torr of helium has a dramatic, negative

effect. Figure A.2 shows the effect of 75 Torr of a mixture of helium and methane, going from pure helium to pure methane. We can see that the organic gas is much better for photoelectron collection. In general, we found that organic gases have much better efficiency than noble gases, the difference being quite large. These results are consistent with measurements made with gas-filled photodiodes*).

We would like to give a simplified explanation of the effect that we see. The atoms of noble gases, having many fewer excitation states available at low energies, are like hard balls scattering the electrons elastically, with a high probability that they will be scattered back into the photocathodes and lost. The electrons have inelastic collisions with the organic molecules, and thus have a smaller probability of being scattered back into the pull of their image charges on the cathodes and lost. But whatever the explanation, the message is clear: for efficient collection of photoelectrons, use an organic gas at as low a pressure as possible.

*) G. Charpak, W. Dominik, F. Sauli and S. Majewski, The gas photodiode as a possible large-area photon detector, IEEE Trans. Nucl. Sci. NS-30 (1983), in press.

Figure captions

- Fig. 1a : Schematic of test detector.
- Fig. 1b : Detail of electrode structure of test detector.
- Fig. 2 : Timing resolution as a function of gas pressure measured for an isobutane-filled low-pressure proportional counter. (Courtesy of Breskin, Tserruya and Zwang, ref. 7).
- Fig. 3 : Emission spectra of BaF_2 measured by Laval et al. (ref. 9), and the transmission of 27 mm of BaF_2 .
- Fig. 4 : Electron component of signal. Time scale is 4 ns/division.
- Fig. 5 : Ion drift of signal. Time scale is 0.5 μs /division.
- Fig. 6 : Timing spectrum measured with 350 MeV α -particles. The time between peaks is 2 ns.
- Fig. 7 : Pulse-height spectrum for 940 MeV protons depositing about 18 MeV in the BaF_2 . The energy resolution is 28.5% FWHM.
- Fig. 8 : The three components of electrons from cosmic rays with the TMAE at 2 °C. The time scale is 50 ns/division.
- Fig. 9 : The three components of electrons from cosmic rays with the TMAE at 20 °C. The time scale is 50 ns/division.
- Fig. 10 : Single component of electrons from the BaF_2 surface after 12 hours of evacuation. The time scale is 50 ns/division.
- Fig. A.1 : Relative collection efficiency of photoelectrons as a function of collection voltage for various pressures of helium. All measurements are normalized to the vacuum measurement.
- Fig. A.2 : Relative collection efficiency of photoelectrons as a function of collection voltage for helium/methane mixtures at 75 Torr. All measurements are normalized to the vacuum measurement.

TEST DETECTOR

DETAIL OF TEST DETECTOR

Fig. 1

Fig. 2

EMISSION SPECTRUM OF BaF₂

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig. 10

Fig. A.1

Fig. A.2