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CsI and Some New Photocathodes

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Abstract

A discussion of the possible sources of discrepancies in the measurements of the quantum efficiency of CsI photocathodes is presented. We propose that the major causes for disagreements in QE are due to the QE dependence on the current density extracted from the photocathode, on the electric field, and on the temperature of the photocathode. Preliminary results on TMAE enhanced GaAs and Si, plus TMAE protected CsTe and SbCs photocathodes, operated in gas, are also presented.

1 Introduction

The last few years have seen a great deal of effort and growing understanding of the preparation and operation of CsI photocathodes coupled to wire chambers, and recently operation in noble liquids [1]. It is well known that photocathodes in a non-vacuum environment are affected by many parameters such as preparation and starting materials, surface layer effects, and electric field strength. Although we have made well over one hundred photocathodes, we still see substantial differences (on the order of a factor of 2) between photocathodes made in "exactly" the same way and with the same starting material. Therefore, it is not surprising that there are differences between authors as to the importance of various parameters, and in particular, disagreements on the absolute quantum efficiency, QE, of a CsI photocathode.

Here we will try to address systematically the sources of the discrepancies, but unfortunately presently the whole story is not "known". What is presented here is part of ongoing work and is the best of our knowledge at the time of this conference. It is likely that at least some aspects of our view will change in the months to come.

Fig. 1 shows the QE of a CsI photocathode as a function of wavelength. Our measurements made by photon counting are shown as well as a measurement made at low electric field, E , in the current mode. A compilation of the measurements made by other authors is also included [2-9]. These data include measurements made by photon counting and in the current mode. The range in measured values of QE varies by factors greater than 2.

Photocathodes that can be coupled to a wire chamber and having substantial QE at wavelengths longer than 220 nm have also been a subject of continuous interest and investigation. Prior to this work, TMAE vapor or condensed TMAE [10] had the lowest threshold for a practical photocathode. It was also discovered that an adsorbed layer of TMAE on some photocathodes makes them more stable in air and even increases their QE [4].

Si and GaAs photocathodes, when covered by a thin film of Cs, form a double layer which makes them sensitive to visible light. Unfortunately, the Cs layer oxidizes when exposed to air. The similarity of the results with Cs and TMAE led us to study the effect of an adsorbed TMAE layer on some conventional photocathodes: GaAs, Si, CsTe, and SbCs. These results will also be presented in this work.

2 QE measurement methods

Most measurements of the QE of CsI photocathodes have been made by current measurements at low electric fields[4, 7, 8, 11]. TMAE gas and calibrated photomultipliers

are used as references. Others have estimated the QE by the number of photoelectrons detected from a Cherenkov radiator [2, 5, 9].

Our QE measurements were made with a vacuum chamber similar to the one shown schematically in fig. 2. The QE of the photocathodes is compared to the QE of TMAE gas using the single-photon technique[12] with a high electric field applied to the photocathode. The chamber is filled with 20 Torr of methane or ethane and the anode-mesh voltage adjusted so that the gain is sufficient to give a well-defined single-photoelectron spectrum and the counting rate is on a fairly-well defined plateau with a slight slope. The pulsed UV light source is filtered by a monochromator and its intensity is reduced by adjusting the input and output slit widths until the number of photoelectrons produced for wavelengths ≤ 190 nm averages between 0.1 and 0.3 per UV pulse (trigger). The number of photoelectrons per pulse is then recorded as a function of wavelength. The pulse rate (trigger rate) is about 120 Hz so any photon or ion feedback from the amplification of one photoelectron will not be counted as a second event due to the approximate 8 ms between 10 μ s wide gates.

The chamber is then refilled with a mixture of TMAE gas and 20 Torr of methane or ethane. The region between the quartz window and the "Anode Mesh" (fig. 2) is operated as a conversion and drift region. For this measurement, the photocathode is held at a higher positive potential than the "Anode Mesh" and used as the true anode. This second region is again operated with sufficient gain to obtain a well resolved single-photoelectron spectrum. For this measurement the counting plateau is very well defined, and the signal has no component from the photocathode. The number of photoelectrons detected per UV pulse is then recorded as a function of wavelength. The QE of the photocathode relative to the TMAE gas is simply the ratio of the number of photoelectrons per pulse for the two configurations. The absorption length and QE of the TMAE gas is taken from the literature[13], with corrections made for the transmission of the grids and for the finite absorption in the space between the quartz window and the anode. This yields the absolute QE of the photocathode. A similar technique was used by Lu et al. [3].

3 Photocathode preparation

Some of the discrepancies seen by various investigators must be due to the quality of the CsI photocathodes. While we have made many photocathodes and still see unexplained variations, we do feel that the starting material and the deposition techniques used are important.

As has been discussed in an earlier publication [14], we have found a significant difference in final QE with different starting materials. We have investigated photocathodes

made from pieces of CsI-scintillator material and from 99.999% pure powder. Vacuum deposition and spray deposition by dissolving in an alcohol and water solution have also been studied. The results of the measurements can be seen in fig. 3. The QE curves are shown for photocathodes made from scintillator (Sc) and powder (P), vacuum deposited (V), and by spray deposited (Sp).

The spray deposited scintillator material and the powder material, both vacuum and spray deposited, had about 70% of the QE of the vacuum deposited scintillator material. A similar result has been seen with photocathodes that have been made from vacuum deposited scintillator and then left exposed to air for a long time. We feel that this difference is due to water absorption by the CsI that cannot be removed by the low temperature heating in a chamber but is removed in the crystal growing process. A much smaller improvement when using scintillator material has also been seen by others [3, 7].

The techniques used for both spray deposited and vacuum deposited photocathodes are well documented in an earlier work [14]. The point to stress is that it is important to first boil off some of the starting CsI material, with the substrate either removed from the system or protected by a shutter. This we believed removes surface and more volatile contaminations and makes a substantial effect in practice. We do not know how important it is but our substrates are held at 40 °C during the deposition of the CsI.

One lesson that has been learned by us and by others, but not yet found in the literature, is that copper substrates are to be avoided. There is some kind of reaction of CsI that has been deposited on copper. The exposure to light seems to be a key ingredient for the reaction. Speculation as to the cause usually centers around the presence of light liberating iodine which reacts with the copper, or a reaction with a small amount of moisture in the chamber. We have found no differences between aluminum or stainless steel substrates photocathodes, either polished or roughened. Some of the best results reported in the literature have been obtained with CsI photocathodes which have been deposited on new aluminum surfaces [4, 15].

4 Optimizing QE

Since the QE of a CsI photocathode depends on the gas or vacuum environment, the electric field strength, and on both the bulk and surface characteristics, it is not surprising that there is not a consensus on how to optimize the QE of a photocathode. Here we will discuss some of these parameters.

4.1 Heating and flushing with gas

For photocathodes that have had prolonged exposures to air, or that have been produced by the spray technique, we consider heating to be the most important ingredient in a good QE [16]. This has been confirmed by some [7, 8], but not seen by others[17]. In this last case the photocathode was heated by putting the entire chamber in a heated enclosure. Our best results have been achieved with heating in a vacuum with the chamber at a lower temperature than the photocathode. Heating has been shown to be less important when the photocathode is made in a good environment and the contact with air is minimized.

There is some disagreement about the effectiveness of improving the QE of a CsI photocathode by flushing with clean gas. This was first reported as an effective technique by Séguinot et al. [4] who flushed with methane at a temperature of 50 °C [18]. Improvement has not been seen by some who worked at room temperature[7, 8]. The effect has also been confirmed by Imre et al. [20], and by Peskov [19] while working at CERN whose photocathode QE increased by a factor of more than two but over a period of several days.

Our CsI photocathodes are typically exposed to air for less than 5 minutes. When heated at 50 °C in a vacuum the QE typically drops by 10% over a period of 4 or 5 hours and then increases to about 75% the starting value over the next day. The initial decrease is believed to be due to the removal of the adsorbed materials from the surface which enhance the QE. The increase in QE is due to the cleaning of the CsI, probably water and absorbed gasses.

There have been communications, both verbal and in the literature that a reduction in QE was seen with heating. In our experience, this is always due to an increase in outgassing from the construction materials condensing out on the photocathode. Often heating is done by heating the entire chamber. In order to prevent condensation of unwanted materials on the photocathode it is important that the photocathode is heated first and maintained at a temperature higher than the environment.

The operating temperature of a CsI photocathode also affects the QE. On this there is also some disagreement. As examples, Malamud et al. [7] see an increase in QE with temperature, but with an optimum temperature of 50 °C, above which the QE decreases. Piuz et al.[17] see a decrease in QE with heating. Again, from our experience, a decrease in QE with heating is the result of outgassing of materials in the chamber at the higher temperatures.

The effect of temperature on the QE of a CsI photocathode depends strongly on the properties of the surface. This is illustrated in fig. 4 which shows the current from a CsI

photocathode in 400 Torr of methane as a function of time for a variety of temperatures and for gains of 1 and 2. The chamber was sealed and the photocathode was under continuous illumination. During the first 21 hours (gain = 1), the temperature was cycled in 10 °C increments from 20 °C to 60 °C and back down to 40 °C, holding each temperature for 3 hours. There was little or no temperature effect seen.

The voltage was then increased to give a gain of 5 for three hours at 40 °C (data not shown) the photocathode was then cooled to 20 °C, and the gain reduced to 2. The temperature was then cycled (data shown) from 20 °C to 60 °C and back down to 20 °C, again holding each temperature for 3 hours. Now the temperature dependence is clearly visible. We believe that the difference between the two sets of data is that the photocathode was cleaned by the return flux of positive ions. This effect of positive ions has been seen by others [21] and attributed to a surface cleansing effect. It has been shown that ion bombardment is an effective way to change the surface layer on photocathodes and in particular for the removal of water [21]. The continued improvement in QE at the higher temperatures on the cool down side of the gain = 2 data is very likely due to continued cleaning of the photocathode.

4.2 Effect of electric field on QE

Although the effect of electric field strength, E , on the QE of CsI photocathodes has been disputed[8], it is well established that the QE increases with E for photocathodes in general [22, 23], and certainly for CsI photocathodes [4, 7, 22, 24]. The dependence of the QE of CsI photocathodes has been measured over a large range of E in a vacuum, but only a small range has been measured in gas due to the onset of amplification. Fig. 5 shows, for several pressures of methane, p , the current as a function of voltage for a CsI photocathode illuminated with 185 nm light. A more revealing representation of the data is presented in fig. 6, where the current is plotted as a function of E/p . It is clear that at the same E/p a higher pressure corresponds to a higher QE. In both figures the dependence of the QE on the applied voltage for a given pressure is clear. In fig. 6 we have included a fit to the linear portion of the 20 Torr methane curve. This fit would imply that the QE at $E/P=19$ would be 50% higher than for an E/p of 2. With the exception of Lu et al. [3] all other measurements have been made with a low E/p on the photocathode. Although the effect of E on QE is significant, it is not enough to explain alone the QE discrepancies among the different investigators.

4.3 Other surface effects

Along with the surface and bulk cleanliness of the photocathode, the gas environment also plays a role in the QE and adds to the controversy. We find that methane and ethane improves the response over that of the photocathode in a vacuum [24]. Braüning et al.[8] see an increase with methane while Malamud et al.[7] see a reduction in the QE. We find that the introduction of methane can increase the QE by as little as 10% to as much as 100%. The difference is probably due to the cleanliness of the photocathode, since it certainly is a surface effect.

We have seen that isobutane reduces the QE of a photocathode over that of vacuum [24]. Piuz et al. [17] sees a 15% reduction in the QE when only 3% isobutane is added to the methane in their chamber. These are good examples of the magnitude of the effect of a contaminating layer on the surface of a photocathode.

We have seen that when a photocathode surface is contaminated, as in the case of operating with isobutane, there is a suppression of the QE at longer wavelengths. This shape is also seen in the measurements of Braüning et al. [8] (fig. 1 curve L) and possibly explains their low QE.

We have also found that a photocathode that is operated with gain in ethane or methane and with an extracted current on the order of a few nA/cm² exhibit a further long-term increase of 20% or more over a period of a day. Fig. 7 shows this effect for a CsI photocathode operated in 100 Torr of ethane and with a gain of greater than 10³. The current increases by about 30% during the first 18 hours. After that the enhancement was dominated by the aging of the photocathode. This increase in QE is stable and is probably explained by the creation of F-centers and ion cleaning of the surface [25].

4.5 Current effects

We have often seen an initial change in the QE of a photocathode due to the current from the photocathode. This has also been seen by others [7, 8, 19]. No point has been made of this in the literature because it was considered a stabilization of the system before the measurement. On closer examination we find that for photon counting it is typical for the counting rate to increase by more than 25% during the first few minutes of exposure to the light source. In the current mode, with current densities on the order of 1 nA/cm² we see a similar sized decrease on the same time scale. We also find that the stabilized current from the chamber is not linear with light intensity. This is called the "fatigue effect" and is particularly applicable for vacuum photodiode where the currents are often large. It is recommended that currents be kept below 10⁻⁸ amp/cm² [21]. This phenomena may well be the missing clue to understanding the result of others.

5 Discussion of CsI results

With the material discussed above and with a closer look at the data presented in fig. 1 we feel that we are able to suggest explanations for many of the discrepancies between the different measurements. Our results measured by photon counting (curve A) and in the current mode (point b) are in excellent agreement with the results of Aleksan et al. made in the current mode (point d) and by counting Cherenkov photons (point c). We believe that the difference in the results of the two modes are due to differences in the value of E and in the currents from the photocathode. The small difference between their point c and our curve A could easily be attributed to the fact that they had a lower electric field on their photocathode. The measurements made by Lu et al. (curve F) were made in the same manner as our curve A and represent either a disagreement in calibration or a real difference in the photocathodes. The points made by Staric et al. (point g) and Arnold et al. (point m) were both made by photon counting at fairly low electric field on the photocathode.

The measurement made by Séguinot et al. is higher than one would expect for a current mode measurement at low E . Their current was extremely low ($\approx 0.8 \text{ pA/mm}^2$) [19] and they operated at 50°C which also enhances the quantum efficiency. The measurement made by Montermann et al. (point h) was also made in the current mode but at extremely low photon flux ($10^2/\text{s}\cdot\text{mm}^2$).

The point measured by Arnold et al. (point m) was obtained by photon counting at fairly low electric field on the photocathode. It is in fairly good agreement with the other high-QE measurements. The measurement of Staric et al. (point g), also made by photon counting at a fairly low electric field, is somewhat lower than the highest values reported. Their results are still nearly a factor of two higher the lower current-mode measurements.

The results of Malamud et al. (curve K) and Braüning et al. (curve L) were made at low E/p and with a substantial current. Here the difference with the higher results can probably be attributed to the low E/p , higher current, and in the case of curve L contamination on the photocathode.

6 Other Photocathodes

GaAs, Si, CsTe, and SbCs photocathodes are sensitive to much longer wavelength than CsI and are widely used in photon detectors, for example vacuum photomultiplier tubes. We try to incorporate these attractive photocathodes in gaseous detectors. It is not an easy task because they are very sensitive to oxygen, water and some other gases and some other contamination. To overcome their difficulties we tried to cover these photocathodes

with a "dominating" layer which protects the cathodes from the influences of other molecules. In this section we present our preliminary results in this direction.

6.1 GaAs and Si

GaAs and Si were originally used for the detection of VUV photons [21]. The crystals were cleaved and used only in a ultra-high vacuum (10^{-9} Torr). In more "practical" vacuum 10^{-5} - 10^{-6} Torr they were found to be unstable because of contamination of the surfaces by gas molecules and eventually were abandoned. It was later discovered that when the clean surfaces were coated with metallic Cs, they became stable and sensitive in the visible. The reason for this is that the Cs gives one electron to the substrate lattice and forms a double electrical level on the surface. The highest QE was obtained with p-doped GaAs and Si treated with Cs. Unfortunately, Cs is air sensitive and the photocathode can be operated only in a vacuum 10^{-6} or less. On the other hand, it is known that some molecules, due to the chemisorption process, also form a double electrical layer on some surfaces [26, 27]. We studied the practical consequences of this effect in more detail.

We tested both n- and p-type GaAs and Si in the experimental setup shown in fig. 2 in low electric fields. In this work, the GaAs and Si used were wafers like those used by the electronics industry. Wafers with different of carrier concentrations from 10^{17} - 3×10^{19} cm^{-3} were tested. The photocathodes QE's were determined by comparison to commercial phototubes and to TMAE vapor.

It was found that both GaAs and Si were initially sensitive to UV light in the range of CsI sensitivity ($\lambda < 230$ nm). The QE went to essentially zero after prolonged exposure to vacuum. The introduction of any gas restored some sensitivity. The best results were found when the clean GaAs and Si wafers were exposed to TMAE vapor. The results are shown in Table I where the QE of CsI at low E/p are also shown for comparison. One can see that all four photocathodes are good at 185 nm and p-type GaAs is equivalent to CsI at that wavelength. At 220 nm p-type GaAs is by far the best photocathode. Exposure of clean GaAs and Si photocathodes to methane or air gave QE approximately 1/20 to 1/30 of those seen for exposures of the materials to TMAE. When exposed to light no variation of the QE in time (contrary to the case of CsI) were found. The results were extremely reproducible and this makes us to feel that these are indeed practical photocathodes.

The photocathodes are extremely stable not only in TMAE, but also in a mixture of TMAE with methane (few Torr to 1 atm). We have checked that in the parallel plate mode of amplification such photocathodes can achieve stable operation with gas gains of up to 10^6 .

Another important feature of strongly p- and n-doped GaAs and Si (concentrations 10^{18} - 10^{19} cm⁻³) is that they are conductive at liquid argon temperatures. This may find an application for the detection of the scintillation light from noble liquids [1]. We have had success with the detection of the scintillation light from liquid argon with GaAs and a systematic study is in progress.

6.2 CsTe and SbCs

Success with GaAs and Si covered by a layer of TMAE encourages us to test other fragile photocathodes such as CsTe and SbCs. Earlier studies of these photocathodes in gaseous detectors were done by Charpak et al [28]. It was found that in the presence of gas the QE always degraded and the speed of this process is proportional to the amount of contaminants (water, oxygen, etc..) in the gas. A thick layer of ethylferrocene or organometallic compounds played an effective protective role and the cathode could be exposed to air. Nevertheless good stability in time was not achieved.

In the study of the effects of TMAE on CsTe and SbCs photocathodes glass envelopes of Hamamatsu vacuum photodiodes were broken in the presence of methane and TMAE vapor by the method described elsewhere [28]. Originally the QEs were around 10% for CsTe (at 250 nm) and for SbCs (at 436 nm) [29]. After the exposure to the gas mixture, the QE dropped to 0.5-1% during the first 24 hours. The QE then stabilized and remained unchanged for a few weeks. The QE at 436 nm as a function of time is shown for a SbCs-TMAE photocathode in fig. 8. This long term stability is much better than obtained by Charpak et al. CsTe photocathodes could be exposed to air without any further degradation in QE. These photocathodes could be operated in the presence of gas with a gain on the order of 10^3 . These preliminary results lead one to consider the possibility of using these TMAE coated photocathodes in an application where the longer wavelength sensitivity is desirable.

7 Conclusion

There is still not a complete understanding of the sources of discrepancies in QE measured for the CsI photocathode. Electric field strength, and current from the photocathode seem to be the largest factors in determining the QE. All the high QE values have been measured by photon counting or with very low photon fluxes. Since the photoelectric effect is basically a surface effect the field will always be plagued by discrepancies and reproducibility problems. But even if one takes the most pessimistic QE from the literature the fact still remains that is a viable photocathode for RICH detectors.

Though there are still disagreements on the details of the CsI photocathode, there is no disagreement that it is now a maturing technique to be used in instrumentation. But, the maturation of CsI, does not signal the end of useful research on photocathodes for use with wire chambers.

We have introduced two new photocathodes, p-type GaAs-TMAE and p-type Si-TMAE that could be operated in wire chambers and that compare very favorably with CsI. P-type GaAs-TMAE has the highest QE of any such photocathode at 220 nm, which is of interest for the detection of the fast component of BaF₂. These deserve much more study.

The protective effects on CsTe and SbCs photocathodes is very interesting and may lead to the development of photocathodes that work in the visible. In particular, the results of the SbCs-TMAE photocathode at 436 nm is very encouraging. A photocathode that is sensitive down to this wavelength and able to operate in a wire chamber would be useful for the detection of scintillation light from a large variety of heavy scintillators.

Table I: QE of p- and n-types of GaAs and Si coated with TMAE and of a CsI at low electric field.

| Wavelength (nm) | GaAs | | Si | | CsI low E/p (%) [†] |
|--------------------|---------------|---------------|---------------|---------------|------------------------------------|
| | p-type (%) | n-type (%) | p-type (%) | n-type (%) | |
| 185 | 10.1 | — | 6.5 | 4.0 | 9.3 |
| 220 | 0.49 | 0.3 | 0.23 | 0.03 | 0 |
| 235 | 0.01 | 0 | 0 | 0 | 0 |

[†]ref. [7]

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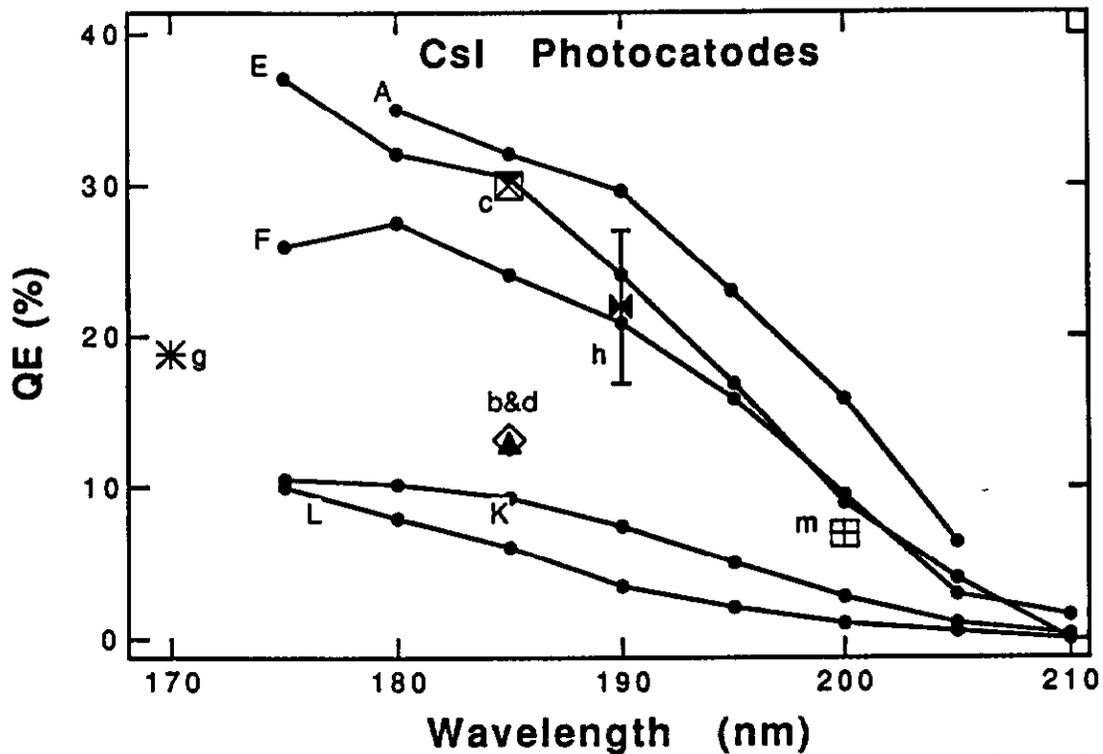


Fig. 1 Quantum efficiency of various CsI photocathodes as a function of wavelength (lower case letters for single points). A: photon counting [this work], b: current mode [this work], c: photon counting [2], d: current mode [2], E: current mode at 50 °C [4], F: photon counting [3], g: photon counting [5], h: low-current mode [6], K: current mode [7], L: current mode [8], and m: photon counting [9].

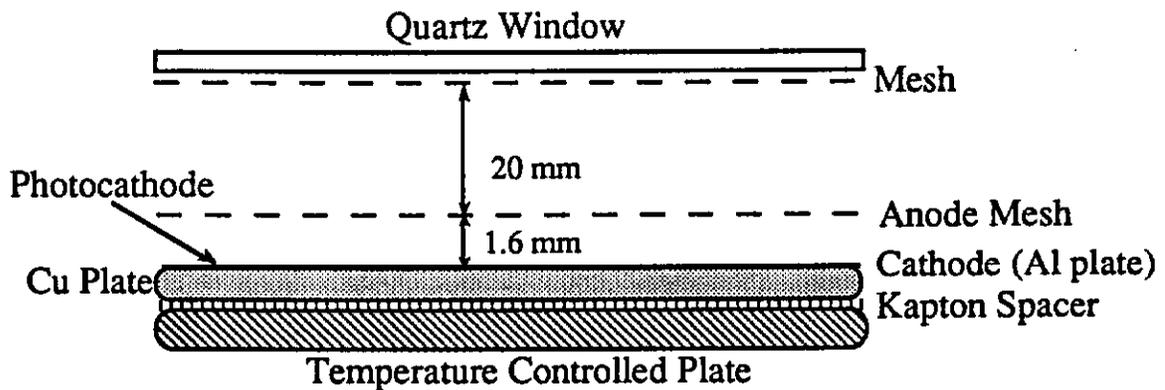


Fig. 2 Schematic of experimental setup.

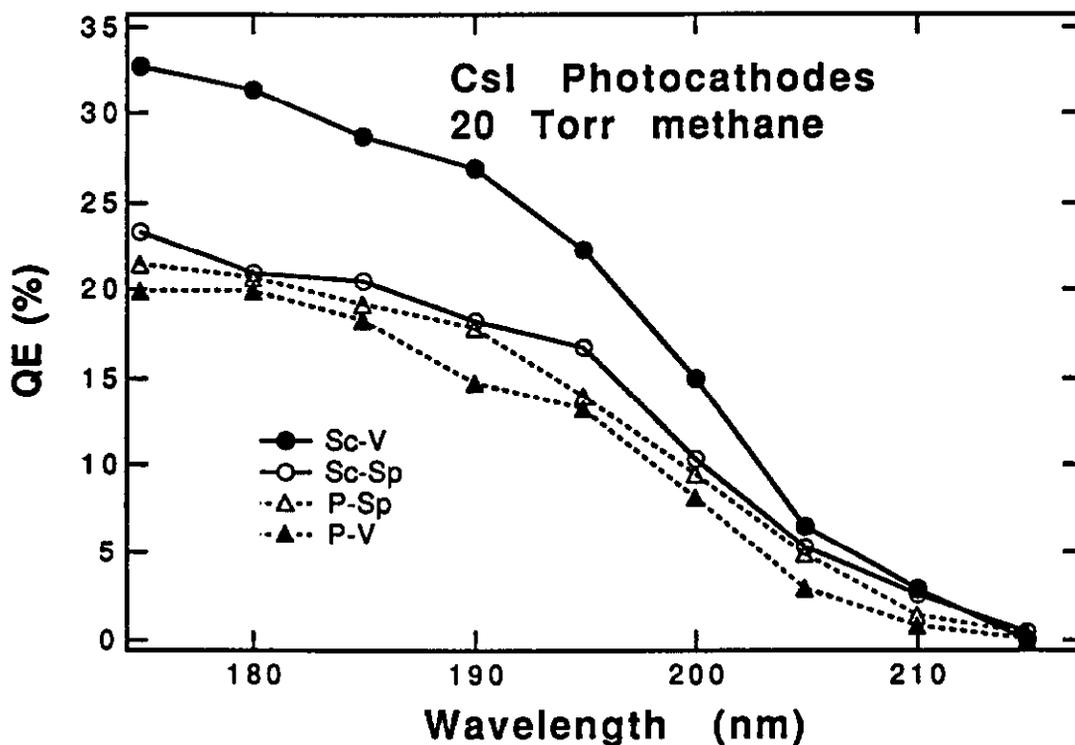


Fig. 3 QE as a function of wavelength, measured in methane, for CsI photocathodes produced from scintillator (Sc) and powder (P) both vacuum deposited (V) and spray deposited (Sp).

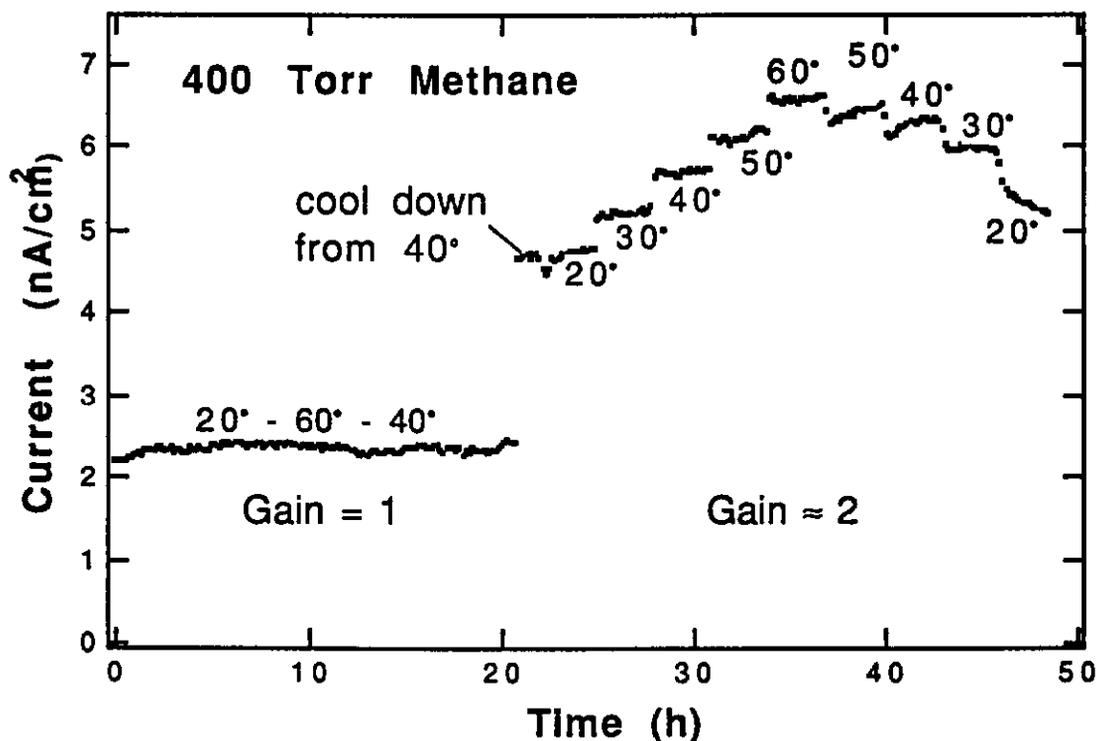


Fig. 4 Current as a function of time for a CsI photocathode in 400 Torr of methane at a variety of temperatures ($^{\circ}$ C) and at gain = 1 and gain = 2. See text.

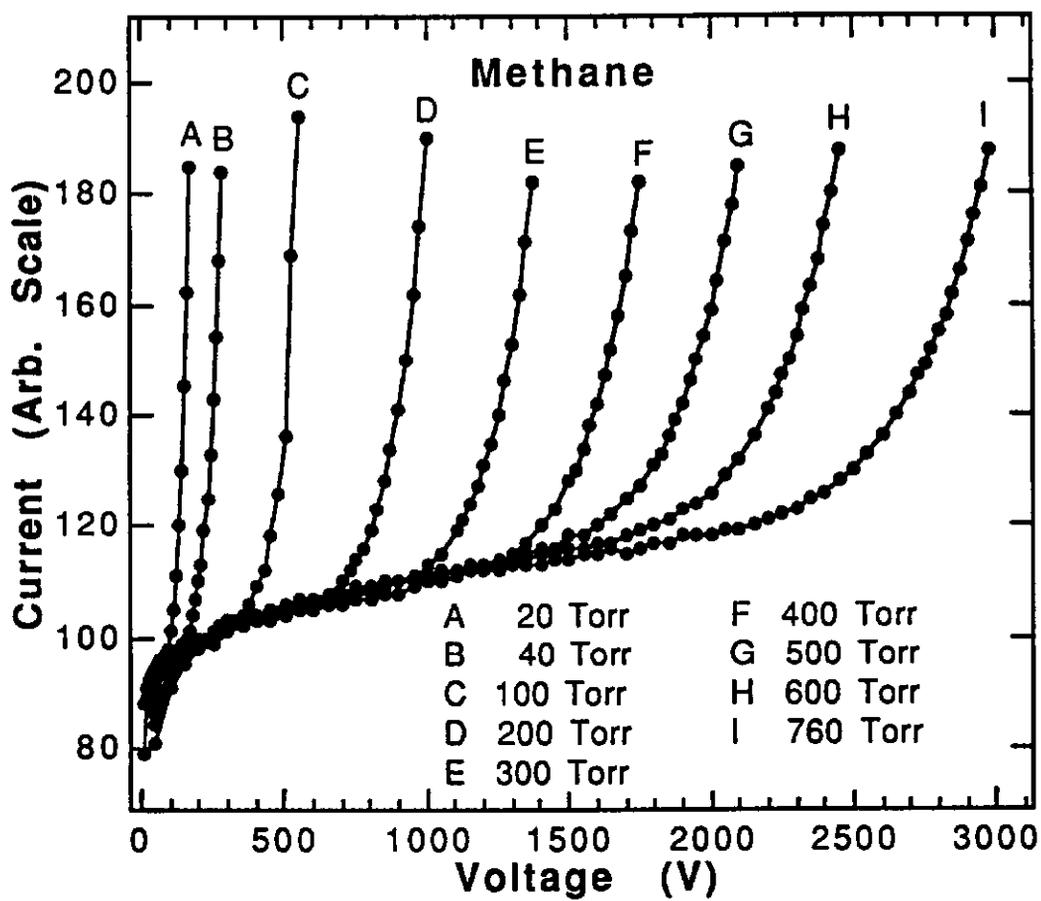


Fig. 5 Current as a function of voltage for a CsI photocathode in a variety of methane pressures.

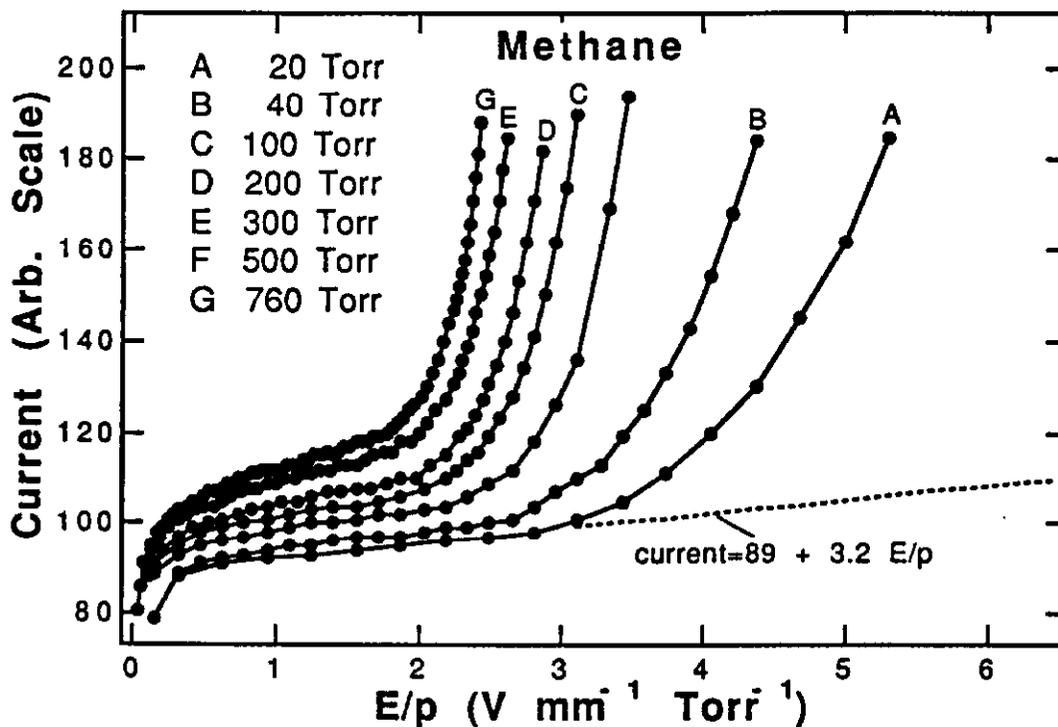


Fig. 6 Current as a function of E/p for a CsI photocathode in a variety of methane pressures.

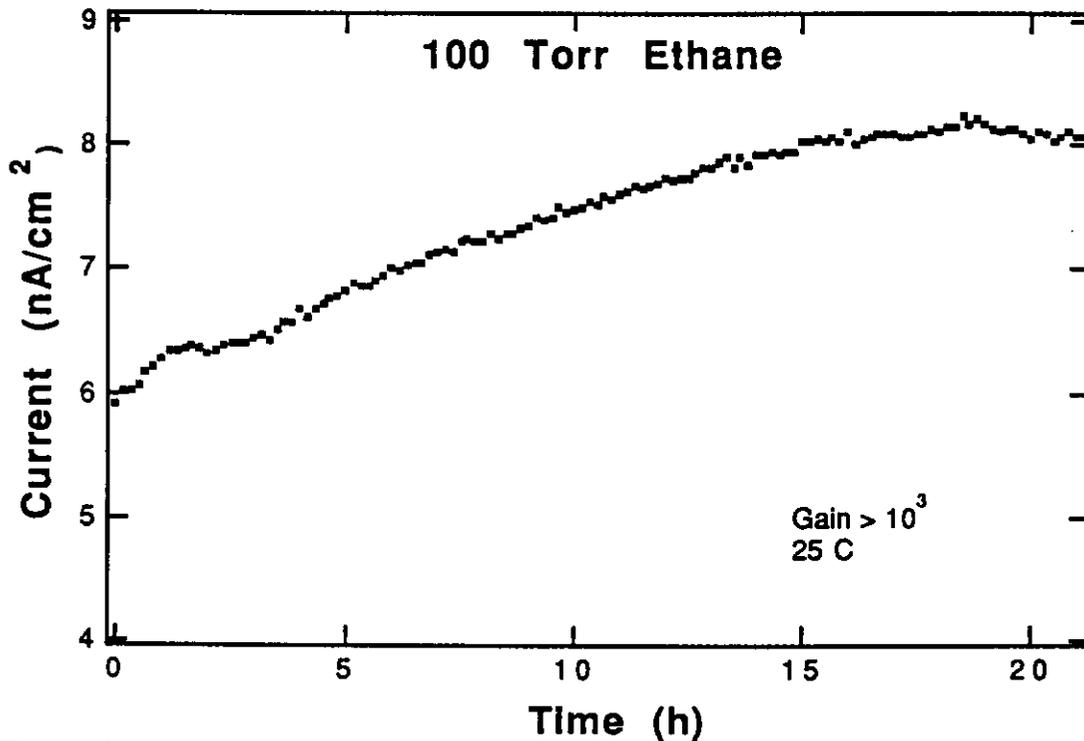


Fig. 7 Current as a function of time for a CsI photocathode operated at 25 °C, in 100 Torr of ethane, and with a gain of $>10^3$.

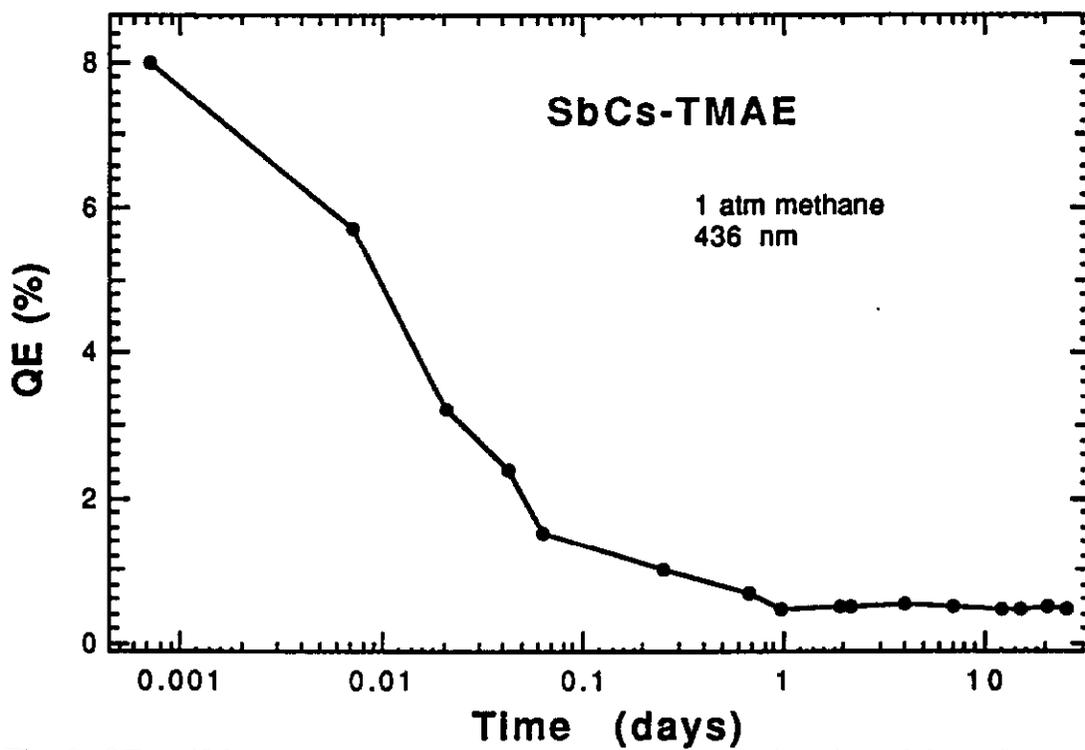


Fig. 8 QE at 436 nm for a SbCs-TMAE photocathode as a function of time after exposure to TMAE and 1 atm of methane.