Advances in Single-Charge Detectors and Their Applications

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Abstract

Modern gas avalanche detectors are instruments of choice for detecting single charges deposited in gas or emitted from thin solid radiation converters. We discuss principal factors governing the operation of gas avalanche photomultipliers, combining solid photocathodes with advanced micro-pattern gaseous multipliers and summarize the properties of UV photocathodes and film-protected photocathodes for the visible spectral range. We review recent progress and applications of single-charge counting detectors and discuss in some detail their application to nanodosimetry and its relevance to studies of radiation damage to DNA.

I. INTRODUCTION

In recent years, gaseous detectors, the traditional workhorse of the detection field, have seen new developments mainly due to micro-lithographic and micro-machining production technologies [1,2]. This results in better detector performance and a variety of new applications. In the present paper we review some advances in single-charge detection in relation to single-photon imaging on one hand and to charge-counting applications on the other hand. Most of the topics have already been discussed in detail in referred publications. Therefore, in this article we will mainly discuss the physical concepts, summarize principal performance and review present and potential applications of single-charge detectors.

II. IMAGING GASEOUS PHOTOMULTIPLIERS

Gaseous detectors, having charge-gains exceeding $10^5-10^6$, are the natural choice for the detection of single photoelectrons. Their combination with a solid-film photoconvertor [3] (which ensures parallax-free response and isochronous electron emission) permits the conception of large-area flat detectors, with single photon localization sensitivity, capable of operation under high photon flux and with very small sensitivity to intense magnetic fields [4]. Similar to vacuum-based devices, the photoconvertor response is the main factor that defines the relevant spectral range.

In the UV spectral range (figure 1) CsI is the best known and employed photocathode, having high quantum efficiency (QE) [5] and being relatively stable under short exposure to air. It is currently incorporated in large-area (square meters) single-photon gaseous photomultipliers, for particle identification by the Ring Imaging Cherenkov (RICH) technique [6]. Other photocathodes of interest could be CsBr [7] and CVD diamond [8]; the latter is chemically stable, radiation hard and can operate under high temperatures. Its

Figure 1. Typical quantum efficiency spectra (in vacuum) of annealed Cs [5] annealed CsBr [7] and hydrogenated CVD diamond [8] photocathodes.

Figure 2. a) Typical absolute quantum efficiency spectra of K-Cs-Sb photocathodes, bare and coated with 300 thick CsBr and 250 thick CsI films. b) The evolution of the absolute quantum efficiency of K-Cs-Sb photocathodes exposed to oxygen. Shown are the results at 312 nm for bare and coated photocathodes (200 and 250 CsI) as function of the residual oxygen pressure. Each data point represents 5 minutes of exposure to oxygen followed by quantum efficiency measurement in vacuum.

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QE, though lower compared to CsI and CsBr, can be further enhanced by proper surface treatment [9], which may be the basis for very good solar-blind photomultipliers.

In the visible spectral range it is more difficult to operate photocathodes under gaseous multiplication; photocathodes in this range (alkali-antimonides or others) are extremely reactive with minute amounts of moisture, oxygen and other impurities of counting gases. A possible solution is to coat the photocathode with a thin protective film [10], which prevents surface chemical reaction with gas impurities at the expense of an attenuation of the photoemission. It has recently been shown that indeed such a coating process can be practiced on alkali-antimonide photocathodes (Cs$_3$Sb, K-Cs-Sb) [11,12,13]. By properly optimizing the coating material and thickness it is possible to maintain the QE value above 5% at a wavelength of 350nm, even under prolonged exposure to oxygen (figure 2). A 250-Å coated K-Cs-Sb photocathode was found to be stable for over an hour in 150 torr of oxygen. The preferred protective coatings are CsI and CsBr, having good electron-transmission properties [13]; due to their hygroscopic nature, however, they were found to be ineffective in protecting the visible photocathodes against moisture. For that purpose further investigations are under way, with hydrophobic organic films.

Another solution to the operation of visible photocathodes within gas avalanche photomultipliers is to use pure noble gas

Figure 3. Fast single electron pulses obtained from a) a Microdot gas avalanche chamber operated in 60 Torr of C$_3$H$_8$; b) A multiwire proportional chamber (MWPC) operated in 40 Torr of i-C$_4$H$_{10}$ and c) the same MWPC coupled to a GEM (at gain 250). All pulses measured with the same fast current amplifier.

Figure 4. Photon feedback in 40-torr Methane: Top) A MWPC coupled to a CsI photocathode exhibits an intense photon feedback, gain=10$^5$. Bottom) A GEM (gain=1) inserted between the MWPC and the photocathode reduces the photon feedback, gain=3x10$^5$.

Figure 5. The multi-GEM photomultiplier concept: 3 GEMs in cascade are coupled to a photocathode. Each GEM operates at a low gain, resulting in a high total gain. The avalanche-induced pulses are recorded on a printed-circuit board.
mixtures. The implementation of this approach is not simple because gaseous electron multipliers normally do not exhibit sufficient gain when operated with noble gases. As will be discussed below, recent developments in gaseous electron multipliers do permit such operation.

In general, the coupling of a gaseous electron multiplier with a solid photocathode involves some particular considerations. The effect of photoelectron backscattering in the gas, reducing the effective QE, can be minimized by a proper choice of the gas type and of the electric field at the photocathode surface [14]. Molecular gases, in particular Methane, are preferable. Applying a high electric field at the photocathode surface can reduce the large backscattering effect in noble-gas mixtures [14]. Of major concern are the secondary avalanche-products, namely photons and ions. Upon interaction with the photocathode they cause severe limitations to the detector gain and localization capability and affect the photocathode lifetime. A careful choice of the electron-multiplier geometry should be made so as to limit photon- and ion-feedback effects. The worst choice would be a parallel-plate electron multiplier, where a high field is applied between the photocathode and an anode electrode; all avalanche-induced ions sputter the photocathode at high velocity, and the avalanche photons initiate secondary avalanches from the photocathode, thus limiting the gain. Contemporary large-area CsI-based photon detectors employ multiwire proportional chambers (MWPCs) [6], in which the ions have high velocity at the vicinity of the anode wires, but they slow down at the photocathode region. Nevertheless, these detectors are not free from photon-induced feedback effects, which limit the gain and the localization resolution. Multi-step avalanche chambers, where the avalanche process occurs in successive multiplication elements, would be a better choice; they offer high gain and only a fraction of the avalanche-ions return to the photocathode [15,16]. Micro-pattern gas electron multipliers such as microstrip, microgap and microdot detectors have been tested in combination with CsI photocathodes at normal and low pressure [17,18,19]. Due to their particular electric field geometry, a large fraction of the avalanche ions are collected at neighboring cathodes on the detector electrode substrate, rather than returning to the photocathode. The rapid ion clearance from the avalanche region results in high counting-rate capability and in fast electrical pulse buildup, both being of prime importance. However, just as in the other electron multipliers, the photocathode is fully exposed to the avalanche-induced photons. Fast multiplication and possibly limited ion-feedback can be reached in other novel multipliers, such as micromegas [20], micro-CAT [21] and GEM (gas electron multiplier) [22], in which the avalanche development is confined to very small distances of typically 50-100 microns. Some examples of fast pulses of single photoelectrons multiplied in some low-pressure micro-pattern detectors are shown in figure 3.

The GEM could become the multiplier of choice for gas avalanche photomultipliers, due to its very special electrode geometry. This simple multiplier consists of a compact array of small apertures in a metal-coated, 50-micron thick, Kapton foil. The apertures, of 50-100 micron in diameter, are typically spaced by 150-200 microns. A potential difference of a few hundred volts across the GEM foil strongly focuses the electrons into the apertures, while an avalanche is developed under a strong and confined electric field, reaching amplification factors above 1000 in a single element, both at atmospheric [23] and low [24,25] gas pressures. Very high gains, exceeding 10^5, and stable operation have been reached in GEM-based photomultipliers (with a CsI photocathode), by cascading a GEM with another multiplying element [23,24] or by cascading several GEMs [23,24]. The GEM has the unique property of having a good efficiency for collecting photoelectrons from a photocathode and focussing them into

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**Figure 6.** Gain vs. voltage characteristics of a 3-GEM photomultiplier with a CsI photocathode, in different gas mixtures. The implementation of this approach is not simple because gaseous electron multipliers normally do not exhibit sufficient gain when operated with noble gases. As will be discussed below, recent developments in gaseous electron multipliers do permit such operation.

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the apertures (defined as transmission efficiency) and at the same time being optically opaque and therefore blocking most avalanche photons. A GEM electrode inserted between the photocathode and a following multiplication element will thus play a multiple role: it would efficiently transmit photoelectrons while screening the photocathode from avalanche-induced photons, as shown in Fig. 4. The GEM would block a fraction of the back-drifting ions, preventing photocathode damage [24]; sharing the charge gain between the GEM and a following element will lead to a more stable operation and ensure high total gain and therefore high sensitivity to single photons or single electrons [23]; the thin GEM electrode leads to a very fast avalanche development, resulting in excellent time resolution.

The GEM transmission efficiency is a crucial issue, particularly when detecting single photons or electrons. It depends on the given GEM geometry, the electric fields above and across the GEM and on the gas and pressure. If the field above the GEM, $E_D$, is very low compared to the field $E_G$ across it, there will be an effective electron focussing into the apertures, but some electrons may be lost due to backscattering and diffusion. As $E_D$ increases the backscattering and diffusion are reduced but the focussing effect might be reduced as well. It has been suggested to further increase $E_D$, to values where multiplication starts and where possible losses of photoelectrons could be recovered by the generation of a few secondary electrons in a small multiplication [24]; this idea is presently investigated. Due to these competing effects the transmission efficiency depends on the fine-tuning of all the parameters [26,27]. The subject has been studied both by Monte-Carlo simulations and experimentally, but so far only relative data exist. The simulations indicate that under some conditions single electron efficiencies close to 100% are expected [27]. It should be noted that the operation conditions for optimal electron collection may contradict those for optimal ion blocking. It is therefore advisable to cascade several GEMs, having more degrees of freedom for the optimal choice of their parameters.

A peculiar property of the GEM is a high attainable gain when operating with pure noble gases [28], due to the blocking of long-range photon-mediated secondary effects. By cascading 3 or 4 GEMs coupled to a CsI photocathode (figure 5), charge gains exceeding $10^5$ were demonstrated with noble gases and their non-aging mixtures, (figure 6), sufficient for single electron detection [28,29]. It was also shown that some gas mixtures (Ar+N₂) yield fast (~10 ns wide) signals (figure 7). The noble-gas operated multi-GEM could be the ultimate solution for gas avalanche photomultipliers operating with non-protected photocathodes sensitive in the visible spectral range.

Figure 7. Anode signals detected with a fast amplifier from the 3-GEM photomultiplier, operating with Ar+10%N₂. The signal, ~10 ns wide, results from ~90 primary electrons emitted from a CsI photocathode.

Figure 8. Top) the concept of the soft x-ray detector. Ionization electrons, induced by photons in the gas, drift in a weak electric field towards an avalanche detector, where they are individually multiplied and counted. Middle) an example of an electron pulse-trail induced by a F-K 676 eV photon, as recorded by the digitizer. 20 torr Ar/C₄H₁₀ (20/80), 200 mm drift length. Bottom) recorded data in a 2D-presentation (pulse trail length vs. number of counted electrons), permitting good separation of x-ray lines and detection of photons down to 100 eV. Shown are lines of Be-K 108.5 eV, C-K 278 eV, O-K 525 eV and Al-K 1486 eV.
range. For that purpose one should search for GEMs produced on compatible clean materials such as ceramic or glass.

III. SINGLE CHARGE COUNTING

Small amounts of energy deposited by ionizing radiation or by low-energy photons in gas can be precisely evaluated by counting the number of deposited ionization charges. This method provides better precision than the standard charge integration and multiplication method, especially for a very small number of deposited charges, since the latter is affected by charge-multiplication fluctuations in addition to the Fano statistical fluctuations of the primary ionization process. The electron counting (or rather electron-cluster counting) method has been proposed for relativistic particle identification [30,31] and more recently as a precise tool for studying basic ionization-deposition process in gases, such as the Fano statistical fluctuations and the mean energy for electron-ion pair production [32]. It was shown that the method could be successfully used for soft x-ray spectroscopy, in the spectral range of 100-1000eV, important for example in material analysis for the identification of low-Z elements in scanning electron microscopes [33]. The method was implemented using a low-pressure gas volume, coupled through a very thin window to the electron microscope. Soft x-rays emitted from light elements in the sample traverse the thin window and are stopped in the gas. The resulting electrons are separated by diffusion while drifting, in a low electric field, towards a high-gain electron multiplier, where they are individually multiplied and efficiently detected and counted. Note that the ionization and detection are both performed within the same gas volume. The requirements of the electron multiplier are similar to those discussed previously for gas photomultipliers. High gains, preferably around $10^6$, are requested, free of secondary effects, with very fast signals, allowing efficient separation and reliable counting of individual electrons within the multi-electron pulse trails. Elements down to Be (108 eV) could be identified by this method (figure 8). The method was implemented with traditional low-pressure parallel-plate and multiwire electron multipliers. It was also tested with low-pressure microdot and microstrip multipliers that provide in some conditions about an order of magnitude faster signals [17,19].

The charge-counting method is particularly attractive for the evaluation of small energy deposits in miniature sampling volumes. In this case the ionization deposits are highly fluctuating, considerably deviating from the average. For this purpose we require a method sensitive down to a single charge but equally precise for measuring multiple charges deposited within a miniature volume.

A specific example is the field of nanodosimetry, dealing with the evaluation of radiation impact to nanometric-size tissue volumes, representing DNA segments. The field is motivated by the current understanding [34] that irreversible radiation damage to a living cell occurs when a small segment of the DNA molecule, typically 30 bases long, absorbs a significant energy quantum. Under this condition, and if both strands of the DNA double-helix are damaged, the cellular mechanism fails to correctly repair the damage and the cell is likely to mutate or die. The goal of nanodosimetry is to provide, for any radiation field, data on the distribution of energy deposition within the relevant DNA-segment volume (2nm diameter, 10-20nm long); it is done with a low-pressure
gas-model (Tissue Equivalent Gas), expanding the DNA dimensions by 6 orders of magnitude. With the help of a newly developed radiobiological model [35], and in correlation with biological essays [36] of DNA and cellular radiation damage, this data can be used to predict the lethality of the radiation field and provide vital information for radio-protection as well as for radio-therapy of cancer.

The charge-counting method is thus used to reliably measure the amount of charges (energy) deposited in a mm-size low-pressure (~1torr) gas volume, from a single charge up to a multitude of 10 charges or more. The latter are the rare events responsible for irreparable DNA damage, and their probability should be reliably evaluated. The concept of the nanodosimeter is schematically portrayed in figure 9: the radiation-deposited electrons, or ions, are extracted by an electric field through a small aperture towards an electron multiplier, where they are individually counted. Using detectors with high efficiency for single charges, the single-charge detection probability is practically defined by the single-charge extraction efficiency: the latter is a function of the aperture size and the charge-transport parameters in the gas. Charges created in the vicinity of the aperture have a high detection probability while those created further away have a lower one. The sensitive volume has an elongated, cylindrically symmetric shape; this wall-less sensitive volume ensures the elimination of secondary wall effects and is of prime importance in view of the small amount of charge to be detected.

In the case of the electron-based nanodosimeter [37] the electrons extracted from the sensitive volume are separated by diffusion along their drift towards a low-pressure gaseous electron multiplier. The deposition, drift and detection are all done under the same gas. This imposes limitations on the gas type and pressure, in order to maintain high gain and to avoid quasi-ballistic motion of the electrons, occurring at very low gas pressures, the latter seriously affects electron extraction into the multiplier. Consequently the electron-based nanodosimeter can simulate tissue volumes of 10-20 nm and above, namely of the chromosome size. Recently published data [38] measured with α-particles in 2.3 torr of propane demonstrate the capability of this method.

In the case of the ion-based nanodosimeter [39], the ions are extracted from the sensitive volume, through a double differential pumping system, into vacuum and are accelerated towards a vacuum-based electron multiplier. This approach permits operation with any gas and the pressure can be very low, simulating even sub-nanometer DNA segments. Moreover, due to very small diffusion [40] experienced by the ions, their arrival time at the detector preserves the primary information of their deposition location. This permits a precise mapping of the ionization density within a multi-nanometer sensitive volume, with sub-nanometer precision. It could also permit the precise mapping of the ionization density with respect to the location of the primary radiation track: an information required by radiobiological models. An example of an ion-trail induced by an α-particle in the ion-counting nanodosimeter is shown in figure 10. The data in figure 11 was obtained with 241Am α-particles in 0.7 torr of propane, within a tissue equivalent sensitive volume of about 1nm³. The probability of having ionization clusters of up to 9 ions in this volume was measured, in very good agreement with theoretical calculations based on gas ionization cross-sections and ion transport parameters.

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Modern gas avalanche detectors offer high gain and fast signals, suitable for the detection of single charges under high radiation flux. Single photon detection and imaging over a large area is required in particle physics and in medicine, and can be realized by combining solid photocathodes with advanced gaseous electron multipliers. Such gaseous photomultipliers are currently applied in the UV range, with CsI photocathodes, in RICH detectors. Recent advances in the passivation of alkali-antimode photocathodes may lead to the implementation of the same technique in the visible spectral range. Alternatively, the peculiar capacity of the GEM to offer high gain even with noble gases could certainly permit the incorporation of regular, bare photocathodes within large area imaging gaseous detectors for visible light, provided that GEMs can be made of compatible clean materials. Cascaded GEMs permit the optimization of the detector structure for minimal ion- and photon-feedback and for prolonged photocathode lifetime. This development may revolutionize the field of photon imaging, permitting inexpensive readout of a large variety of scintillators and scintillating fibers, in many fields of applications.

The single-charge counting method was applied for basic studies of ionization process in gases as well as for soft x-ray spectroscopy. It has recently been applied for the evaluation of small and highly fluctuating energy deposits in miniature gas spectroscopy. It has recently been applied for the evaluation of regular, bare photocathodes within large area imaging gaseous detectors for visible light, provided that GEMs can be made of compatible clean materials. Cascaded GEMs permit the optimization of the detector structure for minimal ion- and photon-feedback and for prolonged photocathode lifetime. This development may revolutionize the field of photon imaging, permitting inexpensive readout of a large variety of scintillators and scintillating fibers, in many fields of applications.

The ion-counting nanodosimetry method is appropriate for evaluating radiation damage in other nanometer samples of condensed matter; it may have a particular interest in evaluating radiation damage to advanced nanoelectronics employed at high luminosity accelerators or in space instrumentation.

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