A SEALED, UHV COMPATIBLE, SOFT X-RAY DETECTOR
UTILIZING GAS ELECTRON MULTIPLIERS*

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December, 2009

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A Sealed, UHV Compatible, Soft X-ray Detector Utilizing Gas Electron Multipliers

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Abstract—An advanced soft X-ray detector has been designed and fabricated for use in synchrotron experiments that utilize X-ray absorption spectroscopy in the study a wide range of materials properties. Fluorescence X-rays, in particular C_K at 277eV, are converted in a low pressure gas medium, and charge multiplication occurs in two gas electron multipliers, fabricated in-house from glass reinforced laminate, to enable single photon counting. The detector satisfies a number of demanding characteristics often required in synchrotron environments, such as UHV compatibility, compactness, long-term stability, and energy resolving capability.

I. INTRODUCTION

SOFT X-rays are an important probe in studies of a wide range of materials, and measurement of C_K fluorescence, at 277eV, as well as N_K at 392eV and O_K at 525eV, can be particularly informative. These studies provide fundamental understanding that ultimately improves the quality of catalysts, polymers, lubricants, superconductors, organic electronics and biological materials, to name just a few. In the field of near-edge extended X-ray absorption fine structure (NEXAFS), a beam of soft X-rays is incident on the sample of interest (Fig. 1). The number and energy of photo-electrons and fluorescent photons that are emitted from the sample permit sensitive characterization of its elemental structure and chemical bonding. The ability to sweep the incident photon beam through a range of energies, plus the different depth sensitivities of electron emission and photon emission, make NEXAFS studies a most important tool in materials science. Fluorescent photon detection in the few hundred eV range is challenging because of the small signal amplitude, and additional constraints of UHV compatibility, good solid-angle coverage and long-term stability, present other constraints. We report here on the development of a new sensor for fluorescent photons based on their conversion in low pressure gas, together with electron multiplication using Gas Electron Multipliers (GEMs) [1].

Manuscript received November 20, 2009. This work was supported in part by the U.S. Department of Energy under Contract No. DE-AC02-98CH10886.

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Fig. 1. Diagram illustrating the basics of x-ray absorption spectroscopy. An intense, tunable beam of x-rays is incident on the sample of interest, and sensors very close-by measure electron and photon emission (photo-electrons and fluorescence photons respectively). Of particular interest in the present development work is a fluorescent photon detector suitable for C_K (277eV), N_K (392eV) and O_K (525eV).

II. DETECTOR CONCEPT

The key properties required of the fluorescent photon detector are:

- i) UHV compatibility
- ii) compact enough to fit into “busy” vacuum chamber
- iii) long term stability
- iv) energy resolving capability
- v) high efficiency
- vi) potential for select band-pass operation
- vii) large solid angle coverage

Existing sensors that have been used are proportional counters, CsI coated electron multipliers, and semiconductor drift detectors. Electron multipliers such as Channeltrons and micro-channel plates are available commercially, are relatively easy to use, but have no energy resolving capability and exhibit efficiency that varies rapidly with photon energy. Silicon drift detectors perhaps best satisfy the key requirements, but as yet are not readily available for soft X-ray detection.

Our groups at Brookhaven and NIST have designed and fabricated gas-filled detectors for a number of years, and in fact reported some of the earliest detectors for fluorescence measurements [2, 3]. For the present studies, we have opted to pursue a design based on electron multiplication using GEMs in a low pressure gas. For photon energies of a few hundred

eV, it is easy to obtain high stopping power in an argon based gas for C_K (277eV), N_K (392eV) and O_K (525eV). A key issue is containing the gas, and developing an entrance window that is thin enough to transmit X-rays into the gas, yet does not permit the gas to diffuse into the UHV chamber.

We have been developing GEMs by drilling holes in copper clad insulating material, along the lines pioneered by Amos Breskin for thick GEMs (THGEMS) [4]. Fig. 2 shows a diagram of the detector designed here for fluorescence studies.

Fig. 2. Diagram of internal arrangement of Gas Electron Multiplier (GEM) electrodes, anode, and sealed detector body. GEMs and anode are spaced 2mm apart, each one 200 μm thick and comprising 200 μm diameter holes on a 450 μm pitch.

Photons enter the detector through a 25mm diameter window, composed of 300nm titanium foil, which is supported on a stainless steel disc with holes that represent a 60% open area. Behind the window is a 15mm conversion and drift region, followed by two GEMs operating in series, and an anode that collects the avalanche charge. In terms of bias, the window is ground, with progressively increasing negative bias applied to GEM1, GEM2 and the anode. Signal from the anode is collected by a charge-sensitive front-end preamplifier, followed by shaping (ORTEC 460 delay line amplifier) and pulse height analysis and counting. Voltage is applied via one of the HT feedthroughs, a resistor chain providing individual bias to each electrode.

A. GEMs

Our goal was a fabrication process which could be undertaken in-house. The most commonly used GEMs, copper coated kapton, require a liquid etching technique that we have not yet attempted to develop. We have been intrigued by the THGEM technique [3], particularly as we have high speed drilling capability for regular printed circuit boards. Thus, we chose to investigate GEMs fabricated from Rogers 4000 circuit board material [5], but keeping the thickness to a minimum, thereby minimizing operating voltage. The smallest diameter hole that can be reliably drilled many times over is 200μm, and in order to maintain a hole diameter to depth ratio of around unity (the typical value for many GEMs), the starting material chosen was 200μm thick. Fig. 3 shows a

photograph of the manufactured GEMs (on the right), and macor spacers and supporting rings on the left. Fig. 4 shows a close-up of the holes drilled in the GEMs, highlighting the uniformity, well-defined cylindrical nature of the holes, and etching of copper on the underside.

Fig. 3. Photograph of ceramic spacers and supporting rings (left), and Gas Electron Multipliers plus anode (right). A US quarter, top right, shows scale.

Fig. 4. Close-up photographs of holes in GEM electrodes. a) shows top of GEM electrodes, b) shows bottom of GEM electrodes, where a 50 μm wide annulus has been etched around all holes to increase the surface leakage path between top and bottom electrodes. GEM electrodes are 200 μm thick and comprise 200 μm diameter holes on a 450 μm pitch.

B. Titanium Window

The most common materials for thin-windowed gas detectors are plastics, such as mylar and polypropylene [2, 3]. However, these materials generate a big change in photon transmission at the carbon edge, making analysis and normalization of NEXAFS data problematic. As an alternative, titanium has been used in this application. The window consists of three layers of 100 nm titanium foil, resting on a stainless steel support disc. The individual titanium sheets contain a small number of minute pin-holes, but three layers together essentially represent a total barrier to gas diffusion. Fig. 5(a) shows the assembled window frame, and Fig. 5(b) a close-up of the unstressed titanium.

Fig. 5. Photographs of titanium window. a) shows detector side, with titanium foil facing the reader – sensitive diameter is 25mm. b) shows vacuum side with titanium non-stressed by pressure differential – support is provided by a stainless steel disc with holes of diameter 1.15mm, on a spacing of 1.41mm, representing an open area of approximately 60%.

The fully assembled detector forms a compact device that is UHV compatible at the working gas pressure. Fig. 6(a) shows the full internal assembly and Fig. 6(b) the complete assembly.

Fig. 6. Photographs of detector assembly. a) shows fully assembled GEMs and ceramic spacers (only the top of GEM1 is visible). The blue-colored chips are part of the HT resistor chain, which is grounded via a final connection to the gas pipe. b) shows complete assembly with body and window in place.

III. OPERATIONAL CHARACTERISTICS

Several competing elements contribute to the optimum operating pressure. Obviously, there needs to be adequate stopping power (pressure) for the X-ray energy in question, but reducing the probability of window rupture and minimizing diffusion of gas into the vacuum chamber require a pressure as low as possible. However, it becomes electrically very difficult to operate this electrode configuration below 50

torr, and an efficient gas mixture for C_K X-rays in ~ 15 mm was chosen (30%Ar/70%CH₄). Fig. 7 shows the efficiency of 15mm of 50 torr of this gas, and around the C_K edge, the stopping power is nearly 100%. However, the full efficiency of the detector should include the window transmission: Fig. 7 also shows the titanium transmission, taking into account the 60% open area of the stainless steel support. The product of these two curves (solid purple curve) represents the complete detector efficiency. As a reminder of edge energies and fluorescent line energies, these are shown in Table I. Fig. 8 shows the calculated efficiency for 15mm vs pressure of 30%Ar/70%CH₄. All efficiency/transmission calculations have been performed using the Lawrence Berkeley Laboratory web-resource [6].

TABLE I. ELECTRON BINDING ENERGIES/LINE ENERGIES OF INTEREST

	C_K	N_K	O_K	Ar_L	Ti_L
Electron binding energy (eV)	284	410	543	248	454
Characteristic line (eV)	277	392	525

Fig. 7. Calculated efficiency of detector, as a function of photon energy. Red (even dashed) curve represents fractional absorption in 15mm gas depth only, blue (uneven dashed) curve represents transmission of 0.3μm titanium window plus its support, and purple (solid) curve is the product of the two.

Fig. 8. Calculated efficiency of detector gas only, as a function of pressure, for C_K X-rays in 50 torr of 30%Ar/70%CH₄.

Clearly, at 50 torr and above the efficiency is essentially unity, but below 30 or 40 torr the efficiency begins to fall rapidly – thus 50 torr is a good balance between efficiency and total stress on the window.

At low pressures of a few tens of torr, if the gas gain exceeds a certain threshold, some form of ion-induced feedback occurs that creates operational instability. Fig. 9 illustrates the anode signals waveforms as this instability develops.

Fig. 9. Waveforms from anode as a function of gas gain, with 50 torr of 30%Ar/70%CH₄:

- a) Effective Gain = 1500 (vertical scale 50mV / div)
- b) Effective Gain = 6000 (vertical scale 50mV / div)
- c) Effective Gain = 18000 (note change in vertical scale to 100 mV / div)

In Fig 9(a), the effective GEM gain is ~1500, creating about 15000 electrons on average, marginally clear of the electronic noise of around 1000 e FWHM. At a gain of ~6000, the signal safely exceeds the baseline noise, and generating valid counts above a preset threshold is straightforward. As the gain is increased further, however, small signals begin to appear about 10 μs after the main signal, a time that is consistent with the drift of positive ions from the anode to GEM2. It also appears from the waveform in Fig 9(c) that some level of parallel plate multiplication is occurring in this gap, as seen by the small positive offset during that 10 μs. This phenomenon is not understood, and is receiving further examination. It should be noted that with operating pressure much below 50 torr, it is not possible to achieve any measurable signals before instability occurs. Conversely, above 75 torr, the behavior is stable even for gains of 50,000.

Under the conditions of fig 9(b), the detector is stable and linear, as shown by the analysis of C_K and ⁵⁵Fe spectra in Fig. 10. For C_K X-rays, resolution is reasonable, around 80% FWHM, and shows little dependence on shaping times over the range 0.25 to 2.0 μs.

Fig. 10. Pulse height spectra for C_K and ⁵⁵Fe at a gas gain of about 6000, showing good linearity and energy resolution.

Fig. 11 Pulse height spectra for C_K at a gas gain of 6000, for various shaping times (electronic gain normalized to remove pulse height reduction as time constant decreased). Energy resolution is about 80% FWHM.

IV. NEXAFS MEASUREMENTS

The soft X-ray detector is now installed on beam-line X7A of the NSLS, an X-ray absorption spectroscopy beam-line managed by NIST. Initial testing has been carried out with a set-up similar to that in Fig. 1, with the fluorescent photon yield detector represented by the soft X-ray detector described in this work. Measurements have been made on a variety of NIST standard samples, and Fig. 12 illustrates two such

evaluations.

Fig. 12. Carbon NEXAFS results from NIST standard samples.

The samples consist of carbon modified silicon powder. In these measurements, the incident primary beam from the synchrotron source was swept in energy from 230eV to 350eV, and the count rate registered from the soft X-ray detector was continuously recorded. Fig. 12(a) and 12(b) shows counts as function of beam energy for a sample containing 9% and 0.7% by weight of carbon, respectively. As the beam energy sweeps through the carbon K-edge at 284eV, there is an obvious increase in counts, due to generation of C_K fluorescence. Even when the primary beam energy is below the carbon edge, the detector count rate is non-zero, due mainly to detection of scattered photons from the sample. As expected, the sample containing less carbon produces a smaller fluorescence flux, but in both examples the peak structure in the rising edge at

284eV is an intrinsic feature of C-C bonding in the sample. An increase in counting rate also occurs at the argon L-edge (248eV), due to the increase in the argon absorption cross-section above 284eV, and it is interesting to note that the peak structures that occur at the rising edge at 284eV are also due to fine structure in the argon electron binding energies.

We have determined from several sets of experiments that the detector represents a well-sealed and clean internal environment for the gas mixture. The diagram in Fig. 2 shows that, in the initial design, the gas system was in flow mode. We are now able to evacuate the detector, back-fill with 50 torr of 30%Ar/70%CH₄ from a single gas-line, valve off that line, and run for over a week without any noticeable change in gas gain or energy resolution.

ACKNOWLEDGMENT

We are grateful for the help of Gene von Achen, Ron Angona, Howard Hansen and Ron Ryan (BNL) in designing and fabricating developmental versions of Rogers 4000 GEMs. We thank Zugen Fu and Chernoy Jaye (NIST) for assistance with the NEXAFS measurements. We appreciate the support and encouragement of Veljko Radeka (BNL) during this project.

REFERENCES

- [1] F. Sauli, "GEM: A New Concept for electron Amplification in Gas Detectors", *Nucl. Instrum. & Meth.* A386 (1997) 531-534.
- [2] D.A. Fischer, J. Colbert and J.L. Gland, "Ultrasoft (C, N, O) X-ray fluorescence detection: Proportional counters, focusing multilayer mirrors, and scattered light systematics", *Rev. Sci. Instrum.* 60 (1989) 1596-1602.
- [3] G.C. Smith, A. Krol, and Y.H. Kao, "A Low-pressure, Parallel Plate Avalanche Chamber for Soft X-ray Fluorescence", *Nucl. Instrum. & Meth.* A291 (1990) 135-139.
- [4] A. Breskin, R. Alon, M. Cortesi, R. Chechik, J. Miyamoto, V. Dangendorf, J.M. Maia, and J.M.F. Dos Santos, "A Concise Review of THGEM Detectors", *Nucl. Instrum. & Meth.* A598 (2009) 107-111.
- [5] Rogers Corporation: Rogers 4000 is low out-gassing, low dielectric constant, high frequency circuit material.
- [6] http://henke.lbl.gov/optical_constants/