Sub-keV x-ray spectroscopy using a low-pressure electron counting detector

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ABSTRACT

We present a new technique for sub-keV x-ray spectroscopy over large area, at room temperature. Photon-induced ionization electron trails deposited in a low-pressure gas are efficiently counted by a multistage electron multiplier. The analysis of the number of deposited electrons and the electron trail length, combined with a detailed simulation of the detection process, provide a powerful method for resolving x-ray lines down to 70 eV. Detection efficiency above 40% and energy resolution of 55% (FWHM) were measured for the Be K (108.5 eV) x-ray line. We demonstrate that light elements with an abundance below 1% in composite samples can be quantitatively resolved. We present the experimental technique, analysis method and x-ray spectroscopy results obtained with the particle-induced x-ray emission technique (PIXE) and at a scanning electron microscope (SEM).

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1 Introduction

High resolution, efficient, large area soft x-ray detectors are required in many fields such as material science, x-ray astronomy and atomic physics. A particular interest is in the 0.1-1 keV x-ray energy range, corresponding to characteristic K lines of low-z materials, as well as higher shell lines of heavier materials.

Though Si(Li) and Ge(Li) detectors with thin windows offer considerable advantages, mostly in good energy resolution for energetic x-rays, they suffer from several limitations in the ultrasoft x-ray energy range. Among these limitations are the necessity to work at cryogenic temperature, their low detection efficiency and their small size. The performance of windowless detectors deteriorates with time due to ice build-up on the detector surface. Several novel techniques providing excellent energy resolutions in the sub-keV energy range are currently under study. Among them are silicon pn-junction detectors [1], and x-ray CCDs [2], both operating at liquid nitrogen temperature and having small active area. Superconductive bolometers [3] provide extremely good energy resolution (FWHM of 7 eV at 6 keV) over relatively small active area, but require operation at temperatures of a few tens of mK.

Contrary to solid-state devices, gaseous detectors operate at room temperature and provide large active area. The intrinsic energy resolution of these detectors is inferior to that of solid-state devices due to the larger energy required to create an electron-ion pair. Gaseous proportional counters provide rather poor energy resolution due to the additional contribution of the electron multiplication fluctuations. When operated at low pressure, they provide an improved energy resolution (i.e. 58% (FWHM) for the C K 277 eV line) due to the reduced fluctuations in multiplication resulting from lower charge density in the avalanche [4]. Gas scintillation proportional counters (GSPC) [5, 6] provide superior energy resolution compared to gaseous proportional detectors. However, they lose of their performance due to the reduction of light yield when operating at sub-atmosphere pressure. The latter is required to permit operation with thin windows.

A different approach for ultrasoft x-ray photon detection was proposed by us a few years ago. It is based on counting primary electrons (sub-ionization energy) deposited by the absorbed photon quanta in a low-pressure gas mixture [7, 8]. The induced electrons are separated by diffusion in a low electric field and are individually amplified in a high gain multistage electron multiplier. The pulse trails of fast current signals are recorded and the number of electrons per trail is proportional to the energy of the absorbed x-ray photon. In this mode of operation, the contribution of the statistical fluctuations of the electron multiplication process are eliminated, and an improved energy resolution, approaching the physical limit given by Fano

statistics is expected.

In this article we report on first spectroscopy results measured with a large area, room temperature electron counting detector in a scanning electron microscope (SEM) and using particle-induced x-ray emission (PIXE). We will present the experimental technique and describe our novel and powerful x-ray line analysis method, combining experimental data and full simulations of the x-ray detection process.

2 The experimental method

2.1 The Electron Counting detector

A schematic view of the Electron Counting (EC) detector is shown in fig.1. It is composed of a 200 mm long radiation conversion and electron drift volume coupled to a double-stage parallel plate electron multiplication element. The active area of the detector is 700 mm². The detector is contained in a vacuum chamber having a 0.5μm thick polypropylene window. It is first pumped to a vacuum of 10⁻⁶ torr and then circulated with a gas mixture, usually a hydrocarbon and a noble gas, selected upon energy resolution and x-ray absorption efficiency [9].

The ionization electrons created by an absorbed x-ray photon, the number of which is proportional to the photon energy, slowly drift towards the amplification element. A low drift field is maintained along the conversion volume by two parallel meshes and equally spaced metal rings, connected to a resistor chain. Diffusion in the gas, enhanced by the low pressure operation and by the long drift path, causes the initial electron cloud to expand during the drift process. Therefore, the electrons arrive at the amplification element at different time intervals and are individually multiplied and counted. The single electron multiplication factor is higher than 10⁶, which guarantees high detection efficiency. Single electron avalanche-induced pulses are amplified using a fast (9 ns rise time), high gain current amplifier and are measured by a LeCroy 5100M digitizing oscilloscope. Each of the single photon-induced electron trails (a sequence of single electron pulses induced by one x-ray photon) is transferred to a PC at a rate of up to 40 Hz. In the future, we will use fast electronics which will enable us to acquire data at rates of 10-50 kHz. The primary electron pulses are automatically recognized and counted using an off-line correlation method, described in detail in ref. [10]. An example of an experimental digitized fluorine-K induced electron pulse-trail is shown in fig.2, together with a simulated electron-trail. The Monte-Carlo simulation was developed in order to provide a deeper understanding of the electron detection processes; it is extensively

used for data analysis, as described below.

2.2 The Monte-Carlo simulation

The Monte-Carlo program simulates all the processes occurring in the EC detector, from the the absorption process of an x-ray in the drift volume to the single electron counting and recognition. The program involves the following: x-ray absorption cross sections, primary ionization statistics, electron drift and diffusion, electron losses in the drift volume, amplification statistics, single electron pulse shape and correlation analysis parameters. All parameters, except for the x-ray attenuation length, are ones measured using the EC detector. The number of primary electrons is randomly generated from a Gaussian distribution, with an average number $\bar{n} = E_\gamma/W_i$ and a standard deviation given by $\sigma = (F \cdot \bar{n})^{0.5}$. $\bar{n}$, E_γ and W_i are the average number of primary electrons, photon energy and the mean energy per ion-pair, respectively.

2.3 Detector performance

Typical distributions of the number of counted electrons, (analog to the pulse height distribution in a proportional counter), measured at 20 Torr of Ar/C₂H₆ (20:80) for the two extreme x-ray energies investigated: Be K 108.5 eV and Al K 1486 eV, are shown in fig.3a. The Al K spectrum was measured using a 200 nm Al filter foil which strongly attenuates the oxygen line of the Al₂O₃ target. One may notice the good energy resolution and signal-to-noise ratio of the Be K line. The arrows in the plot indicate the expected average number of counted electrons for Be K and Al K lines, assuming 100% single electron counting efficiency and a selected mean energy required per ion pair, $W_i=26$ eV. They emphasize the known non-linear counting efficiency of the EC technique, dominated by the overlap of electron pulses at large electron densities [7]. One can also notice the carbon line, around 9 electrons, due to carbon build-up on the Be target. An additional effect, also seen in fig.3a, is the tailing of the Al line towards the low number of counted electrons. This is due to photons absorbed deep within the drift volume, resulting in a shorter drift path and therefore lower diffusion, causing a reduction of the counting efficiency. The combined effect of a large photon attenuation length (37 cm for the Al K at 20 Torr of Ar/C₂H₆) and a large number of induced electrons (≈ 60) is well emphasized in the corresponding experimental and simulated contour plots (fig.3 b and c) of the number of counted electrons versus the electron-trail length. Such a tail of short electron trails and smaller number of counted electrons, extending to the axis origin, is seen for Al K photons; at lower energies the effect is much

smaller. As can be clearly seen in this figure, there is a correlation between the x-ray energy to the scatter of these photon events. This correlation enables us to recognize, isolate and remove part or all of different x-ray lines. An improvement of the energy resolution is obtained by selecting, in a controlled way (having good knowledge of the loss of efficiency involved), only part of the events, namely events which were subjected to large diffusion and therefore have large electron counting efficiency. An example of such procedure is shown in fig.4, in which (a) and (b) correspond to the full Al K spectrum, and to the spectrum of events within the window plotted in fig.3 b (experimental) and 3 c (simulated). The tailing of the Al spectrum in fig.4b is completely removed, resulting in a significant improvement in energy resolution from 33% to 24% (FWHM). The solid lines in fig.4 correspond to simulated number of counted electron distributions under the same event selection. The good agreement between the simulated and experimental electron distributions is clearly seen in both cases, pointing out to the high accuracy of our simulation technique.

The x-ray lines analysis method is based on the incorporation of the Monte-Carlo simulation. The simulation provides the x-ray selection windows according to the detector parameters and the photon energy, and permits the calculation of the shape and intensities of various x-ray lines under any combinations of selection windows. The simulation provides us the tools for data analysis using the scatter plot. The simulation is performed once for all the relevant x-ray energy range, for a given counting gas, providing the necessary data base for the experimental data analysis.

3 Results

Characteristic x-ray spectra were measured using both PIXE at our Van de-Graaf accelerator and a SEM. We will describe below two typical cases and demonstrate the power of our analysis tools, combining experimental data and the simulation procedure. The basic algorithm is the identification of x-ray lines by removing all or part of the most intense lines. Then, by fitting the experimental distributions of the number of counted electron to simulated distributions, under various window selections, the intensities of various lines are extracted.

Number of counted electron distributions measured at the SEM using a 7.7 keV electron beam striking a mixed target: O (34.82%), Si (20.4%), Ti (11.58%), Ba (33.21%) and coated with carbon are shown in fig.5. The measured scatter plot and number of electron distribution are shown in fig.5 a and b. The C K (277 eV) and O K (525 eV) lines are easily identified in fig.5b. By excluding 95% of the C K events within the simulated dashed selection window in fig.5a, one can identify x-ray lines

at $\approx 100, 190, 450, 900, 1100$ eV and 1750 eV, pointed out by arrows (fig.5c). By fitting simulated number of counted electron distributions for various x-ray lines to the full experimental distribution, we have identified 11 different lines, 10 of which are below 1.2 keV. The overall χ^2 of the simulated lines fit to the experimental data is 1.01. In order to emphasize the strength of the method, we have selected only part of the events using simulated windows at x-ray energies of 100, 277, 525, 970 and 1750 eV (solid lines in fig.5a). The selected electron distribution is shown in fig.5d. A simulation fit to the electron distribution using only the five chosen energy windows, is not shown in fig.5, but provides a χ^2 of 3. Only by fitting this electron distribution to all the identified energies, one can get a χ^2 of 1.06 (fig.5d). This emphasizes the strength of the window selection method for recognizing x-ray lines, without losing substantial information. The accuracy in determining the x-ray line energy is of the order of 10 eV, while the error in the determination of the line intensity is of the order of 0.2% from the overall number of events.

A summary of the expected lines and their corresponding intensities is given in table.1. With this technique, the two lines at energies of 452 and 458 eV can not be resolved. In addition to the expected lines, we identified two additional lines which were also found in other targets on the same target holder. These may be high shell lines of other materials contaminating the targets surface. It is important to note that we were not affected by bremsstrahlung background using the EC detector, in contrary to a simultaneous measurement using a Si(Li) detector mounted in parallel in the SEM. This may be due to the use of magnets in front of the EC detector, eliminating background resulting of back-scattered electrons, and to the low density of our detector. This lack of bremsstrahlung background is of considerable importance in this field and is subject of systematic investigations.

As a second example, we have combined two sets of data of B (183.5 eV) and Be, C (108.5, 277 eV) lines with different intensities, recorded with the PIXE technique. The original distributions of the number of counted electrons are shown in fig.6 a and b, while the combined scatter plot and the counted electron distribution are shown in fig.6 c and d. By eliminating the B K line from the combined distribution using the boron selection window shown in fig.6 c, one can recognize both the Be and C lines (fig.6e). A simulated fit to the combined electron distribution provides all 3 line intensities (fig.6d). The original and the respective extracted line intensities are as follow: B K 97% and 96.5%, Be K 0.8% and 1.2% and C K 2.2% and 2.3%. It is important to note that part of the C K photons originate from carbon build-up on the boron target. The resolving ability of this technique in the very low-energy range can reach sub percents level in elemental concentrations, both in PIXE and SEM.

4 Conclusions

We have presented a novel large area, room temperature x-ray detector, based on electron counting in low-pressure gas, and an associated x-ray spectroscopy analysis technique. We characterized this detector using both PIXE and SEM techniques. A combined analysis of the number of counted electrons and the electron pulse-trail length enables to resolve close x-ray lines, even when overlapped, and to identify as much as 10 x-ray lines in the energy range of 0.07-1 keV. X-ray line intensities below one percent can be quantified even in a relatively noisy environment such as in PIXE and SEM. Indeed, the low density of the detector makes it practically insensitive to charged particles or more energetic photon background. Compared to other methods employed in surface analysis, this technique provides better detection efficiency and energy resolution in the very low energy range. The use of thin windows is possible due the low-pressure operation, and provides detection efficiencies of tens of percents in the sub-keV energy range over active area of hundreds mm^2 .

The combination of the high resolution electron counting detector and a Monte-Carlo simulation of the detection process, provides a unique and efficient technique for resolving low intensity, very low energy x-ray lines.

A combination of such sub-keV electron counting detector with a Si(Li) detector may provide an excellent solution to x-ray spectroscopy from about 70 eV to tens of keV.

In the near future, we will replace the electron multiplier by a gas microstrip multiplier [11] or other faster devices, providing shorter single electron pulses with improved signal-to-noise characteristics. This should permit improving the energy resolution of the detector, and making more compact devices. Using a three-dimensional readout (2D spatial+electron arrival time) may in principle permit reaching the Fano-limited energy resolution in the x-ray energy range of 0.07-1.5 keV.

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References

- [1] R. Hartman et al. *Nucl. Instr. and Meth.*, A377 (1996) 191.
- [2] R.P. Kraft et al. *Nucl. Instr. and Meth.*, A361 (1995) 372.
- [3] D. McCammon et al. *Nucl. Instr. and Meth.*, A370 (1996) 266, and references therein.
- [4] D. Vartsky, A. Breskin, R. Chechik, M.B. Goldberg and G. Malamud. *Nucl. Instr. and Meth.*, A301 (1991) 96.
- [5] A. Policarpo. *Nucl. Instr. and Meth.*, 118 (1974) 221.
- [6] T.Y. Hamilton, C.J. Hailey, W.H.Ku and R. Novick. *IEEE Trans. Nuc. Sci.*, NS-27 (1980) 191
- [7] A. Pansky, G. Malamud, A. Breskin, and R. Chechik. *Nucl. Instr. and Meth.*, A323 (1992) 294.
- [8] A. Pansky A. Breskin, R. Chechik, and G. Malamud. *Nucl. Instr. and Meth.*, A330 (1993) 150.
- [9] A. Pansky, A. Breskin, and R. Chechik. *J. Appl. Phys.*, 79 (1996) 8892-8898.
- [10] G. Malamud, A. Breskin, and R. Chechik. *Nucl. Instr. and Meth.*, A307 (1991) 83.
- [11] A. Breskin, E. Shefer, R. Chechik and A. Pansky. *Nucl. Instr. and Meth.*, A345 (1994) 205.

Figures captions.

1. A schematic layout of the Electron Counting detector. X-ray induced electron clusters diffuse under drift in a low electric field. Individual electrons are multiplied and counted. Their number is proportional to the x-ray energy.
2. Typical 676 eV, fluorine x-ray events: (a) a digitized experimental electron trail.(b) a simulated electron trail. Electron pulses, recognized by an algorithm are circled. Ar/C₂H₆(20:80) at 20 Torr.
3. 1D and 2D distributions of the counted number of electrons, induced by Be K and Al K photons in Ar/C₂H₆(20:80) at 20 Torr. (a) measured distributions of Be K and Al K ; notice an additional C-K induced peak, around 8-9 electrons. The expected average numbers of counted electrons, assuming 100% efficiency and a W_i value of 26 eV, are indicated by arrows in the figure. Solid lines connecting the experimental points are added to guide the eye. (b) and (c) are experimental and simulated contour plots of the counted number of electrons as a function of the pulse-trail length. The identical selection windows shown in (b) and (c) are used for analysis (see fig.4).
4. Distributions of experimental (squares) and simulation (lines) counted number of electrons. (a) includes all events, while (b) includes only events within the two-dimensional windows shown in fig.3 b and c, for the experimental and simulated data, respectively.
5. Different representations of x-ray fluorescence spectra measured in a scanning electron microscope using a target consisting of oxygen, silicon, titanium, barium, coated by carbon. (a) A 2D scatter plot of the data. (b) Distribution of the number of counted electrons including all events. The solid and dotted lines are simulated x-ray line fit to the data and their sum, respectively. (c) Distribution of the number of counted electrons, after removing 95% of the C K x-ray events using the dotted window in (a). (d) Distribution of the number of counted electrons after selection of events using the solid windows shown in (a).
6. Combined distributions of number of counted electrons in Be and B targets, measured using a PIXE technique in Ar/C₄H₁₀ (20:80) mixture at 20 Torr. (a) and (b) correspond to the two original individually measured spectra of B and Be. The combined scatter plot and distribution of the number of counted electrons are given in (c) and (d), while (e) corresponds to a combined counted electron distribution after removing the intense peak of B K using the B K window shown in (c).

Table caption

1. Expected x-ray lines and measured x-ray line intensities, from a calibration target. The two unidentified lines at the bottom do not appear on the manufacturer's list and are due to contaminations.

Expected x-ray lines (eV)		Measured x-ray lines (eV)	
x-ray line	energy (eV)	energy (eV)	intensity %
Ba N _{IV,V} O _{II,III}	≈ 75	75	1.9%
Si L _{II,III}	92	92	2.9%
C K	277	277	30%
Ti L _{III,M_I}	395	-	-
Ti L _{II,M_I}	401	400	0%
Ti L _{III,M_{IV,V}}	452	450	6.8%
Ti L _{II,M_{IV}}	458	-	-
O K	525	525	17%
Ba M _{IV,N_{III}}	601	600	1.6%
Ba M _{IV,V} ,O _{II,III}	765-789	780	6.3%
Ba M _{III,N_{IV,V}}	973	973	5.3%
Si K,L _{II,III}	1740	1740	11.4%
-	-	≈200	12.7%
-	-	≈1100	4.4%

Figure 1.

Figure 2.

Figure 3

Figure 4

Figure 5

Figure 6.