

Development of low-energy X-ray spectrometry at the Laboratoire National Henri Becquerel

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In the frame of the French Metrology Institute, the Laboratoire National Henri Becquerel performs accurate characterization of semiconductor detectors that are in use in a number of applications. Their efficiency calibration, energy resolution and the detailed shape of their response function are parameters of interest for accurate processing of low-energy X-ray spectra to be applied to elements identification and fundamental research studies. The tools specifically developed for low-energy detectors calibration and characterization are described, from the use of radioactivity standard to the development of a tunable monochromatic X-ray source.

Keywords: X-ray spectrometry; semiconductor detector; calibration; metrology.

El instituto francés de metrología, Laboratoire National Henri Becquerel, realiza la caracterización exacta de los detectores a semiconductor que son utilizados en numerosas funciones. La calibración del rendimiento, la resolución en energía y la forma detallada de la “función respuesta” de estos detectores, son parámetros de interés para el tratamiento exacto de los espectros de rayos X de baja energía. Estos espectros, son aplicados a la identificación de los elementos y a la investigación fundamental. Las herramientas desarrolladas específicamente para calibración y caracterización de los detectores de rayos X de baja energía son descritos: del uso del estándar de la radioactividad al desarrollo de una fuente monocromática de rayos X.

Descriptores: Espectrometría de rayos X; detector a semiconductor; calibración; metrología.

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1. Introduction

Semiconductor detectors are commonly used for low-energy X-ray analysis. They are applied to a large range of studies, from fundamental research applications in various fields, such as environment, biology or archaeometry for example.

In the frame of the French Metrology Institute, the Laboratoire Henri Becquerel (LNHB) must accurately characterize semiconductor detectors that are in use for performing radionuclide activity measurement or photon emission determination. Efficiency, energy resolution and detailed shape of the response function of low-energy X-ray detectors are parameters of interest for accurate processing of low-energy X-ray spectra

Different studies have been conducted to improve the knowledge of detectors characteristics and spectrum processing. This paper gives some examples of the tools that have been developed at LNHB with this objective during the last few years, from the use of radioactivity standard to the development of a tunable monochromatic X-ray source.

2. Difficulties for calibration in the X-ray energy range

In the frame of radionuclide metrology, determination of X-ray emission intensities requires accurate efficiency calibration of the detectors and detailed spectra processing to derive accurate photoelectric peak area and relevant low associated uncertainties.

The commonly used method for efficiency calibration in gamma- and X-ray spectrometry is the use of radioactive standards: knowledge of the radionuclide certified activity

(A) and of its photon emission intensity with energy E, ($I(E)$), allows the computation of the efficiency corresponding to E, $\varepsilon(E)$:

$$\varepsilon(E) = \frac{N(E)}{A \cdot I(E) \cdot t} \quad (1)$$

$N(E)$ is the peak area corresponding to energy E, obtained during the acquisition time, t.

The associated uncertainties depend on the uncertainties on each component: peak area, activity and emission intensity. Indeed, a major difficulty in using standards is the accurate determination of the peak area, $N(E)$, as often several peaks overlap in X-ray spectra. However, even if both first parameters can be known with accuracy better than 0.5%, the X-ray emission intensities are generally poorly known (2-3%), contrarily to gamma rays that have better accuracy (e.g. ^{137}Cs : $I_{\gamma 662 \text{ keV}} = 0.8499$ (20), i.e. relative uncertainty = 0.23%) [1]. As example, Table I presents the emission intensities for ^{51}Cr , ^{55}Fe and ^{57}Co , radionuclides that can be used for calibration in the energy range around 5 keV [1]. Consequently, the relevant efficiency uncertainty directly suffers from this major uncertainty factor and efficiency calibration can hardly be obtained with a standard combined uncertainty better than 2% in the energy range lower than 10 keV.

In return, if the efficiency calibration is used to determine emission intensities, the associated uncertainty should not be better than about 2% at the best. Moreover, in both cases, there are corrective factors, such as standard source self-absorption that can attain up to 10% for ^{55}Fe for a deposited point source, thus increasing the final uncertainty.

TABLE I. Example of uncertainty associated to radionuclide emission intensities.

Radionuclide	Energy	Photon emission intensity	Relative combined uncertainty (%)
^{51}Cr	4.9447 (V $\text{K}_{\alpha 2}$)	6.79 (14)	2.06
	4.95224 (V $\text{K}_{\alpha 1}$)	13.36 (27)	2.02
	5.42735-5.46296 (V $\text{K}_{\beta}^{\text{I}}$)	2.69 (7)	2.60
^{55}Fe	5.8877 (Mn $\text{K}_{\alpha 2}$)	8.45 (14)	1.66
	5.8988 (Mn $\text{K}_{\alpha 1}$)	16.56 (27)	1.63
	6.4905-6.5354 (Mn $\text{K}_{\beta}^{\text{I}}$)	3.40 (7)	2.06
^{57}Co	6.39084 (Fe $\text{K}_{\alpha 2}$)	16.8 (3)	1.79
	6.40384 (Fe $\text{K}_{\alpha 1}$)	33.2 (5)	1.51
	7.05798-7.1081 (Mn $\text{K}_{\beta}^{\text{I}}$)	7.1 (2)	2.82

Thus, there are major difficulties for accurate metrological work in the X-ray energy region, and for most of the applications using low-energy semiconductor detectors, as the efficiency calibration is required for quantitative analysis, the same problems appear.

3. Efficiency calibration using monochromatic beam

In regards of the above-mentioned difficulties for the low X-ray energy region, it was decided to develop another efficiency calibration method, independent of radioactive standards. This was first obtained using a monochromatized beam at the LURE synchrotron facility (Orsay, France). According to Bragg's law, a double-crystal monochromator selects a specific energy in the continuous synchrotron radiation. Different crystals are used (Beryl, InSb, Si, Ge), depending on the required energy. The monochromatic radiation is finely collimated and sent to the semiconductor detector to be calibrated. The photon beam reaches the detector window under a normal angle of incidence.

The efficiency calibration is provided by means of a removable proportional counter that is used to determine the incident photon flux [2-4]. The proportional counter (PC) includes a polymer window and an absorption gas volume (Ar-CH_4). The transmission of the window has been measured and the gas pressure is accurately monitored ((500 (10)) hPa) thus the theoretical efficiency of the PC can be determined with combined standard uncertainty between 1 and 5% with a maximum value around argon K edge.

The resulting full-energy peak efficiency calibration obtained for a silicon-lithium (Si-(Li)) detector is displayed in Fig. 1, where the experimental points are displayed with their uncertainty bars; moreover, by scanning the absorption edge of the main components of the detector (K edge of Si, Al and Ni, L edge of gold), the thickness of these have been determined, thus leading to the efficiency calibration curve with combined standard uncertainty better than 2% in the 1-7 keV energy range.

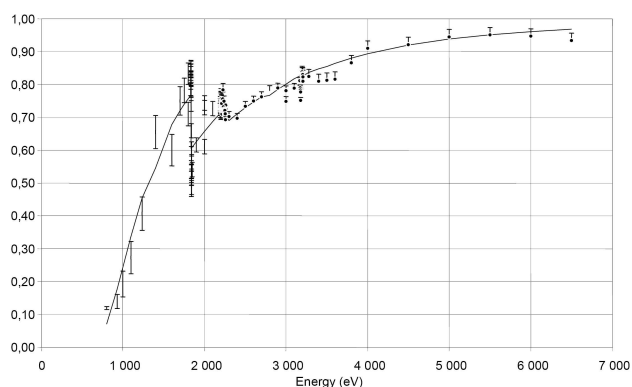


FIGURE 1. Example of full-energy peak efficiency calibration for a Si(Li) detector.

4. Response function calibration

The tunable incident radiation makes possible examining and identifying the different spectrum features by fine scanning around the binding energies of the detector materials, thus improving the knowledge of the detector response function and the subsequent processing of complex X-ray spectra.

The spectrum shape strongly depends on the photons and photoelectrons interactions at the level of the semiconductor crystal-electrode interface. Part of low energy side of the full-energy peak is characterized by features resulting from electron interactions: escape of photoelectrons and Auger electrons from the active part of the crystal and penetration of photoelectrons and Auger electrons of the electrode into the detector active part. Then, the response function is characteristic of a given detector characterized by the active crystal material (generally silicon or germanium) and the one of its electrical contact (gold, nickel, palladium, etc.). The experimental evidence is shown by the spectra shape obtained with a given detector for different photon energies.

4.1. Si(Li) detector

Different "generations" of silicon-lithium detectors have been studied using the monochromatic radiation. One of

the first studied Si(Li) detector has a relatively large crystal (area = 28 mm², thickness = 5 mm), with a gold electrode; its resolution (full width at half maximum) is 170 eV at 5.9 keV and the peak-to-background ratio is rather low (about 1 000 at 5.9 keV) [2]. One of the main results with such an “old” detector is the dramatic shape change observed at the K binding energy of silicon (1.84 keV). Figure 2 shows spectra recorded with incident energy just below (Fig. 2a) and just above (Fig. 2b) this energy.

As soon as the incident energy passes beyond the silicon K binding energy, the primary interaction depth significantly decreases. The large low energy tail appearing above the binding energy is then attributed to incident energy deposition in a silicon “partially active layer”, near the electrode contact, thus resulting in degraded events on the low-energy side of the main peak.

With a more recent detector, this effect of “partially active layer” appears only slightly, but as the spectrum background is significantly reduced, features due to electron escape appear. This is shown with a small Si(Li) detector (area = 10 mm² – thickness = 5 mm) equipped with a nickel electrode. FWHM is 135 eV and the peak-to-background ratio is 15 000. As for the previous detector, just below silicon K binding energy, (Fig. 3a) the main feature of the spectrum is the full energy peak with its Gaussian shape. The low-energy bump should be consecutive to the penetration of L Auger and photoelectrons of the nickel electrode after a

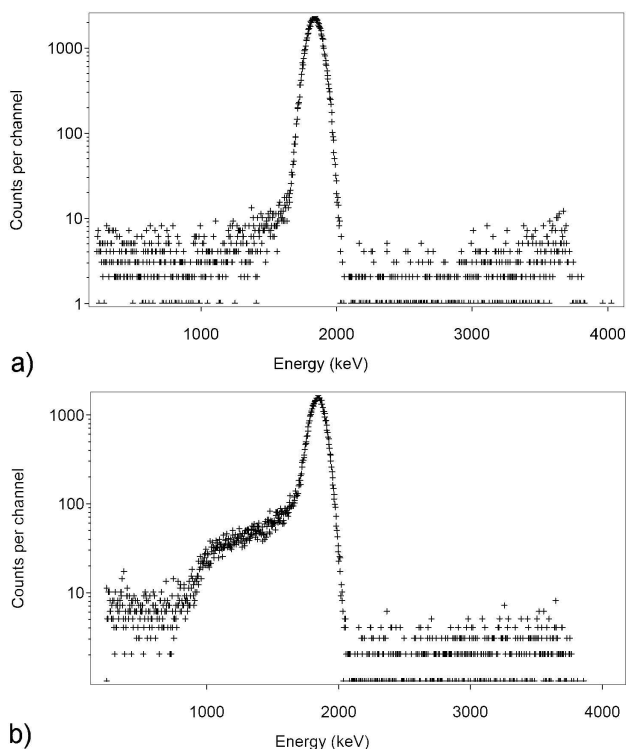


FIGURE 2. a) Spectrum with incident energy 1 832 eV obtained with “Old” Si(Li); b) Spectrum with incident energy 1 838 eV obtained with “Old” Si(Li).

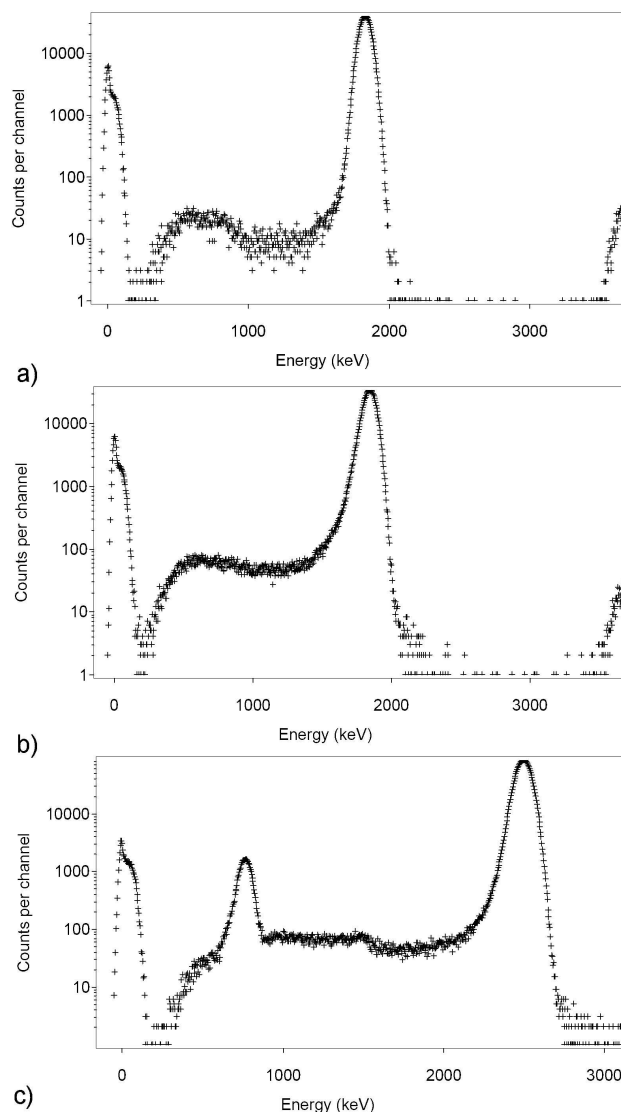


FIGURE 3. a) Spectrum with incident energy 1 810 eV obtained with “New” Si(Li); b) Spectrum with incident energy 1 850 eV obtained with “New” Si(Li); c) Spectrum with incident energy 2 500 eV obtained with “New” Si(Li)

primary photoelectric effect in this part of the detector; these electrons leaving only part of their energy in the detector bulk. Just above this energy (Fig. 3b), the Gaussian shape is enlarged by a slight left-sided tail but the background is enhanced by a factor of about 10: the latter is due to silicon K photoelectrons and Auger electrons escaping from the crystal. With incident energy E , these electron escape effects give continuous energy distributions respectively from $E_K(\text{Si})$ (1.84 keV) and $E - E_{K\text{Auger}}(\text{Si})$ ($E - 1.6$ keV) to E . At last, in Fig. 3c, as the energy increases, the silicon K escape peak appears at $E - E_{XK}(\text{Si})$ ($E - 1.75$ keV). The latter different features are energy evolving according to the incident energy, maintaining a constant energy difference with the main peak energy position.

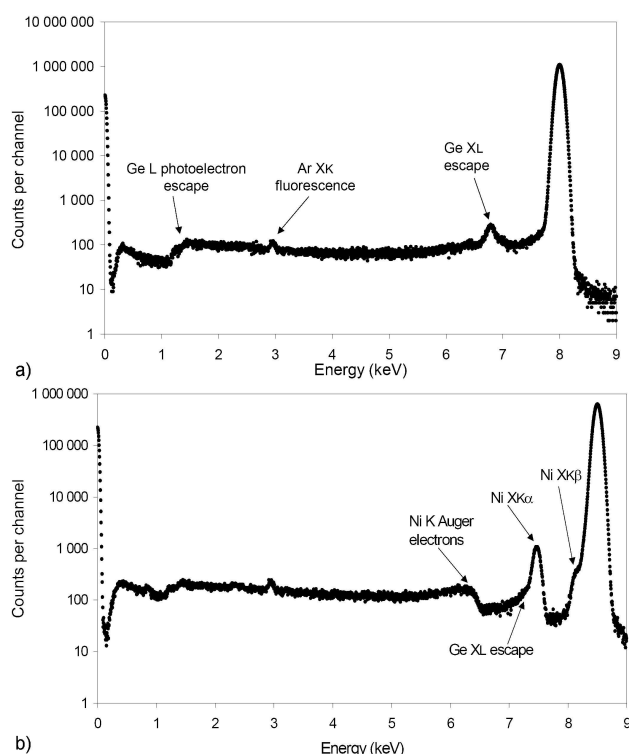


FIGURE 4. a) Spectrum with incident energy 8 keV obtained with HPGe; b) Spectrum with incident energy 8.5 keV obtained with HPGe.

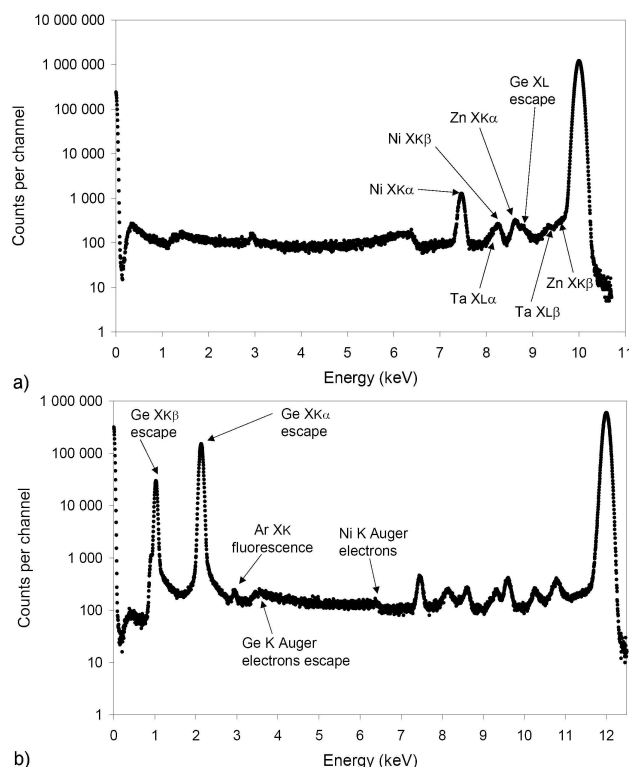


FIGURE 5. a) Spectrum with incident energy 10 keV obtained with HPGe; b) Spectrum with incident energy 12 keV obtained with HPGe.

4.2. HPGe detector

The study of the silicon detector has mainly shown the effect of electrons escape from the detector bulk. Moreover, we studied the response function of a high purity germanium (HPGe) detector with excellent resolution (110 eV at 5.9 keV) and peak-to-background ratio of about 10 000 [4]. Here, the penetration of electrons from the nickel electrode is shown with incident energies selected on both sides of the Ni K binding energy. At 8 keV (Fig. 4a), below the nickel K binding energy, the incident photon interaction occurs with L shell; there is no significant effect in the spectrum. At 8.5 keV (Fig. 4b), the ionization is mainly obtained via photoelectric effect on the K shell inducing Ni K photoelectrons and subsequent electronic rearrangement produces both K photons and Auger electrons. All these secondary particles can enter the detector thus giving either fluorescence peak (photons) or continuous distributions due to the electrons. At higher energy, the effect of interaction in K germanium shell is also clearly demonstrated when incident energies are selected on both sides of Ge K binding energy (11 keV). At 10 keV (Fig. 5a), the spectrum shows the same components as at 8.5 keV. Just above the K binding energy, the primary interaction mainly occurs with Ge K electrons, thus inducing Ge K photoelectrons, and consequent rearrangement gives Ge K photons and Auger electrons. If the primary interaction occurs near the crystal edge, all these particles can escape the active part, thus giving escape effect: the well-know escape peaks, and also continuous distribution due to escape of photo- and Auger electrons. These different effects are shown in Fig. 5b where the spectrum is obtained with 12 keV incident photons.

4.3. Spectra processing

The response function of recent semiconductor detectors used in the low-energy range reveals features that were previously hidden due to high background. Using a monochromatic X-ray radiation, several shapes due to photon and electron interaction were qualitatively identified. Moreover, different experimental studies and Monte Carlo simulations [5-8] confirm the identification of most of the observed elemental components.

To study the response function with accurate description of the peak shape, the COLEGRAM software is used [9-10]. This code has been developed to fit mathematical function to experimental data using the least squares methods, and particularly to process experimental photon spectra obtained with semi-conductor detectors. It is mainly characterized by its ability to model different peak shapes in the same region of interest, and to use individual parameters for each peak, including the natural photon linewidths. Different functions specifically dedicated to the low-energy X-ray range are available. Figure 5 presents an example of the so-called "XLOW" function, adapted to the processing of some spectra in the low-energy range. It includes a basic Gaussian, an exponential tail, a truncated tail and a continuum.

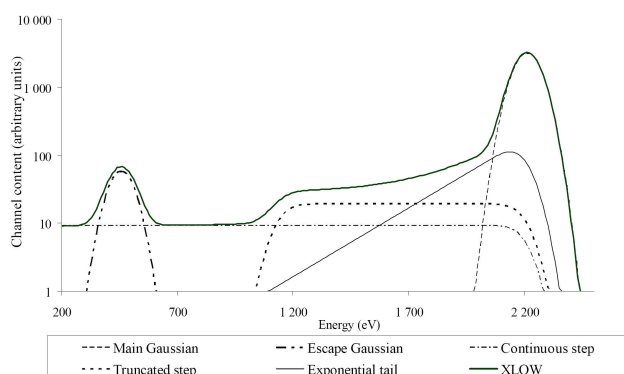


FIGURE 6. Components of the “XLOW” function.

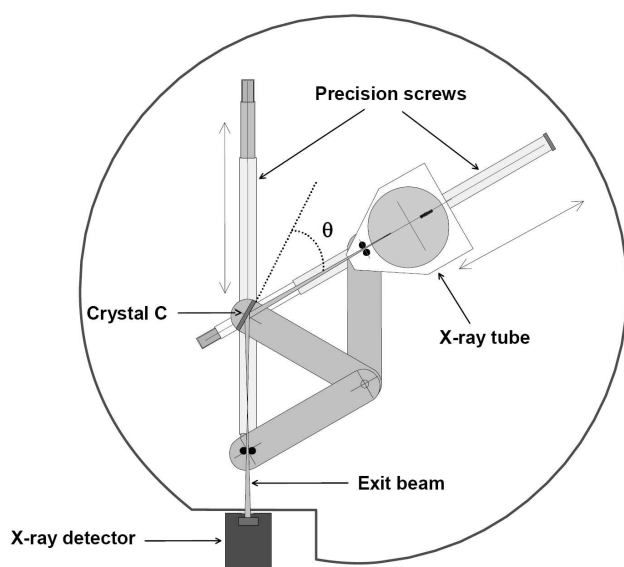


FIGURE 7. Top view of the SOLEX mechanism in the reflection mode. θ is the Bragg angle. C is the centre of the crystal; the vertical axis in C is the rotating axis of the crystal.

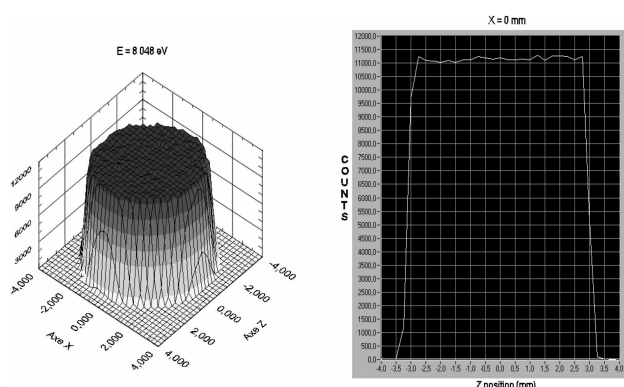


FIGURE 8. Experimental measurement of a HPGe detector surface homogeneity using SOLEX.

5. Development of a monochromatic X-ray source

The studies at the LURE synchrotron facility demonstrated that the use of a monochromatic photon beam, with tunable energy and intensity, allows accurate analysis of the detectors characteristics. It was then decided to provide LNHB with a specific setup to provide monochromatic radiation in the 1–20 keV energy range. This facility, the SOLEX (Source Of Low-Energy X-rays) have been installed in late 2001 at LNHB [11].

It basically includes an X-ray tube and a curved-crystal wavelength dispersive spectrometer both installed on an accurate mechanical system that insures that the monochromatic radiation is produced in a fixed direction. The spectrometer can be switched to either the Johann (reflection mode) or Cauchois (transmission mode) geometry by a simple rotation of the crystal. The whole setup is included in a large circular vacuum chamber. Figure 7 presents the scheme of the SOLEX mechanism. The monochromatic output beam intensity depends on several parameters: X-Ray tube high voltage and current, crystal reflectivity, collimation and selected energy. The emission spectrum of the X-ray tube consists in a continuous spectrum (Bremsstrahlung) whose maximum energy (keV) correspond to the applied high voltage (kV) and anode characteristic X-rays whose intensity is much higher than the Bremsstrahlung one. Thus, the counting rate on the detector ranges between a few tenths of photons per second, when the energy is selected in the Bremsstrahlung continuum, and several thousandths per second when the energy selection is performed at the characteristic X-ray energy of the anode X-ray tube (example: copper K_{α} X-ray at 8.048 keV from the anode tube).

Up-to-now, this source has been applied to relative characterization of detectors, in terms of their response function. SOLEX is also equipped with a scanning system to examine the homogeneity of the detector surface. Figure 8 displays the result of such measurement on a HPGe detector. The scan is performed with a monochromatic beam at 8.046 keV collimated at $350\ \mu\text{m}$, with $250\ \mu\text{m}$ steps in both X and Z directions. For each position the counting rate on the detector (spectrum total area) is recorded. In this case, the detector area shows a homogeneous profile (measurement uncertainty around 1%) and the measured active surface is a circle with about 5.5 mm-diameter.

In the next future, SOLEX will be equipped with a reference detector to provide efficiency calibration, such as it was previously made at the LURE synchrotron. Finally, the monochromatic beam is used for measurement of attenuation coefficients: this has been made for solid materials, such as aluminum and copper [12], and recently for liquid scintillators [13], to optimize the liquid scintillation measurements performed at LNHB.

6. Conclusion

During the last few years, the Laboratoire National Henri Becquerel has conducted studies to improve the knowledge of low-energy semiconductor detectors. Such characterization has been initially performed by means of a monochromatic synchrotron radiation at the LURE synchrotron facility. It was thus demonstrated that the use of a monochromatic photon beam, with tunable energy and intensity, allows accurate analysis of the detectors characteristics. The use of a tunable monochromatic radiation makes possible examining and identifying different features included in a low-energy X-ray spectrum, by fine scanning at the binding energies of the detector materials, thus improving the knowledge of the

detector response function and the subsequent processing of complex X-ray spectra.

As the LURE facility definitely closed on December 2004, the installation of the low-energy tunable X-ray source, SOLEX (Source Of Low-Energy X-rays) in LNHB allowed to continue to perform this kinds of studies. This laboratory setup makes it possible to perform routine studies like in synchrotron facility, with the advantage of high schedule flexibility. Moreover, in the next future, the Laboratoire National Henri Becquerel will be able to work at the SOLEIL synchrotron that will operate its first beamlines in 2006. LNHB is partner of the metrology beam line that will especially dedicated to metrological studies and is planned to start its operation in late 2007.

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