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Use of tunable monochromatic X-ray sources for metrological studies in the low-energy range at the Laboratoire National Henri Becquerel

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Abstract. In the frame of the French Metrology Institute, the Laboratoire National Henri Becquerel (LNHB) performs accurate measurements of photon emission intensities in order to improve the knowledge of radionuclides decay scheme. This is achieved using semiconductor detectors, however, in the low-energy range (E < 20 keV) their efficiency calibration is not straightforward. Moreover, their energy resolution and the detailed shape of their response function are parameters of interest for accurate processing of low-energy X-ray spectra. For these purposes, and in complement to the use of classical calibration method using standard radionuclides, an original method for detectors characterization have been developed using tunable monochromatic radiation. The tunable radiation makes possible examining and identifying the different features of X-ray spectra 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. Since 2001, the SOLEX source (Source Of Low Energy X-rays) has been installed at LNHB and provides tunable monochromatic radiation in the 1–20 keV energy range. This facility has been applied to the measurement of photon emission intensities and attenuation coefficients of materials (Al, Cu) in the low-energy range. In the next future, the SOLEIL synchrotron facility will be equipped with a metrology beam line that will be partially managed by LNHB: this will allow subsequent development of such metrological studies with enhanced performances.

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 determining photon emission intensities. 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 the photon emission intensity associated with energy E, I(E), allows the computation of the efficiency corresponding to E, ε(E):

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

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 know 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_γ (662 keV) = 0.8499(20), i.e., relative uncertainty = 0.23%) [1]. As example, table 1 presents the emission intensities for ^{51}Cr, ^{55}Fe and ^{57}Co, radionuclides that are commonly used for calibration in the energy range around 5 keV. 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.

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 exist.

Table 1. Example of uncertainty associated to radionuclide emission intensities [1].

Radionuclide	Energy (keV)	Photon emission intensity	Relative combined uncertainty (%)
^{51}Cr	4.945 (V $K_{\alpha 2}$)	6.79 (14)	2.06
	4.952 (V $K_{\alpha 1}$)	13.36 (27)	2.02
	5.427–5.463 (V K_{β})	2.69 (7)	2.60
^{55}Fe	5.888 (Mn $K_{\alpha 2}$)	8.45 (14)	1.66
	5.899 (Mn $K_{\alpha 1}$)	16.56 (27)	1.63
	6.491–6.535 (Mn K_{β})	3.40 (7)	2.06
^{57}Co	6.391 (Fe $K_{\alpha 2}$)	16.8 (3)	1.79
	6.404 (Fe $K_{\alpha 1}$)	33.2 (5)	1.51
	7.058–7.108 (Fe K_{β})	7.1 (2)	2.82

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 selected a specific energy in the continuous synchrotron radiation. Different crystals were used (Beryl, InSb, Si, Ge), depending on the required energy. The monochromatic radiation was finely collimated and sent to the semiconductor detector to be calibrated. The photon beam reached the detector window under a normal angle of incidence.

The efficiency calibration was provided by means of a removable proportional counter that was used to determine the incident photon flux [2–4]. The proportional counter (PC) included a polymer window and an absorption gas volume ($Ar\text{-}CH_4$). The transmission of the window had been measured and the gas pressure was accurately monitored (500 (10) hPa) thus the theoretical efficiency of the PC could 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 shown in figure 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.

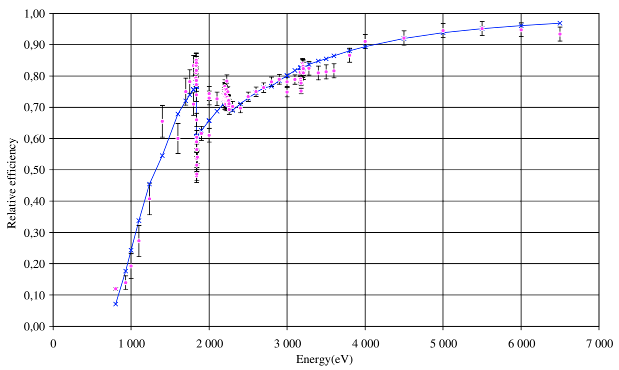

Fig. 1. Example of full-energy peak efficiency calibration for a Si(Li) detector.

4 Response function calibration

The tunable monochromatic 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, etc.). The experimental evidence is shown by the spectra shape obtained with a given detector for different photon energies. As example, figure 2 shows the spectrum shape obtained with a HPGe detector for incident energies just below and above Ge K binding energy, showing the tail increase due to the partially active layer.

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 decide 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) has been installed in late 2001 at LNHB [5]. It basically includes an X-ray tube and a curved-crystal

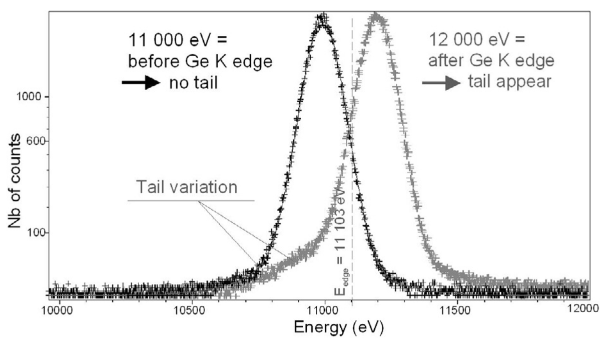

Fig. 2. Comparison of spectra with incident energy 10 keV obtained with HPGe detector.

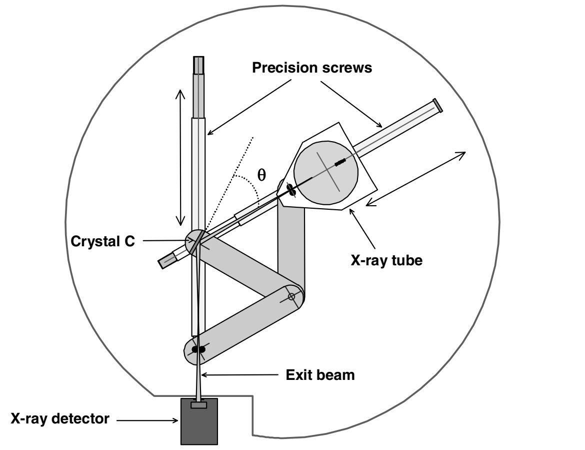

Fig. 3. Top view of the SOLEX mechanism in the reflection mode. θ is the Bragg angle. C is the centre of the crystal; the vertical axis in C is the rotating axis of the crystal.

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 3 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 tens 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. The monochromatic beam is used for measurement of attenuation coefficients: this has been made for solid materials, such as aluminium and cooper [6], and recently for liquid scintillators [7], to optimize the liquid scintillation measurements performed at LNHB.

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.

6 Absolute efficiency calibration

Moreover, recently, the efficiency calibration of an HPGe detector was derived directly from the characterization of its components constituting absorbing layers between the incident radiation and the germanium active layer [8].

In this study, SOLEX was used to determine the thickness of detector components by an energy scanning around their K or L binding energies, according to the following steps:

A monochromatic beam with intensity I_0 and energy E, normally incident through material i, is absorbed according to Beer-Lambert's law:

$$I = I_0\, exp\left(-\frac{\mu_i(E)}{\rho_i}\cdot\rho_i\cdot x_i\right) \quad (2)$$

with

I: transmitted beam intensity,
x_i: material thickness (cm),
ρ_i: material density (g.cm^{-3})
$\mu_i(E)/\rho_i$: mass attenuation coefficient (cm$^2\cdot$g^{-1}, function of the energy).

At energies close to binding energies of any material, the mass attenuation coefficient presents large variations [9].

Thus, the transmitted photon beam and the relevant counting rates below and above the edge, respectively $I_{below\ edge}$, and $I_{above\ edge}$, show a discontinuity the amplitude of which, A_{edge}, allows determination of the material thickness:

$$A_{edge} = \frac{I_{below\ edge}}{I_{above\ edge}} = \frac{exp\left(-\frac{\mu_{below\ edge}}{\rho}\cdot\rho\cdot x\right)}{exp\left(-\frac{\mu_{above\ edge}}{\rho}\cdot\rho\cdot x\right)} \quad (3)$$

where $\mu_{\text{below edge}}/\rho$ and $\mu_{\text{above edge}}/\rho$ are the mass attenuation coefficients of the material at energies below and above the binding energy, respectively. From equation (3), the thickness can be determined as:

$$x = \frac{ln(A_{edge})}{\left(\frac{\mu_{above\ edge}}{\rho} - \frac{\mu_{below\ edge}}{\rho}\right)\cdot\rho} = \frac{ln(A_{edge})}{\Delta\mu\cdot\rho} \quad (4)$$

$$\text{where } \Delta\mu = \left(\frac{\mu_{above\ edge}}{\rho} - \frac{\mu_{below\ edge}}{\rho}\right).$$

Thus, for each energy step around the studied edge, the detector energy spectrum is recorded and the variation of the full-energy peak (FEP) area is used to determine the edge amplitude.

Using this procedure, the layers of aluminium (infrared shield), nickel (electrical contact) and germanium (dead layer) have been measured. The sensitivity of the method makes the measurement of very thin thicknesses possible (a few tens of nm with a precision of a few percent); indeed it revealed the presence of a germanium dead layer and allows measuring its 12 nm-thickness. Moreover, complementary spectra processing allowed characterization of the partially active layer. This information is fundamental when characterizing the electrode-germanium crystal interface and for the knowledge of the intrinsic characteristics of semiconductor detectors.

This approach ensured accurate determination of the thickness of each component and made possible the detector efficiency calculation by Monte Carlo simulation. Differences between simulated data and experimental efficiency values (obtained from standard radionuclides and from a previous experiment using reference beam from LURE synchrotron facility) were about 1% to 5% for energies above 1 400 eV, what reveals an excellent agreement.

7 A beam line dedicated to metrology at SOLEIL

SOLEIL is the new French synchrotron facility installed in Saclay; it is a source of the third generation type, characterized by a synchrotron light emission all along the storage ring circumference. Both a research laboratory and a very high-level scientific resource serving thousands of users, SOLEIL has just started to operate in late 2006.

The metrology beamline is conceived with the objective of providing a calibration and metrology test facility for detectors and optical components. This will be installed on a bending magnet and will include three branches respectively dedicated to VUV, soft X-rays and hard X-rays energy ranges to cover 10 eV to 15 keV. The beamline is designed to provide great flexibility to first address the needs of the metrological studies and to be used as a general-purpose beamline to prepare, test and set up a wide range of experiments.

The Laboratoire National Henri Becquerel is partner of this metrology beam line that is planned to start its operation in early 2008. This will be the opportunity to have a monochromatic photon beam at our disposal, with high photon flux dynamic: this will allow, for example, using narrowly collimated beam (some tens of nm) to get high accuracy on the detectors characterization.

8 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 continuing to perform these 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 very next future, the Laboratoire National Henri Becquerel will be able to conduct the same types of experiments at SOLEIL, with more details, taking advantage of the powerful quality of the synchrotron source.

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