CH9700039

Prompt Gamma-ray Activation Analysis (PGAA)

Jean Kern

Physics Department, University, CH-1700 Fribourg, Switzerland

1. Introduction

There are in principle two ways to use neutrons for elemental and isotopic abundance analysis in samples. One is the "neutron activation analysis", a method presented here by Prof. Gäggeler. I will label it as "off-line", since the neutron-induced radioactivity is observed after the end of the irradiation. The other method, which I will call "on-line", consists in observing the capture gamma-ray during neutron bombardment. This will be the subject of the present lecture.

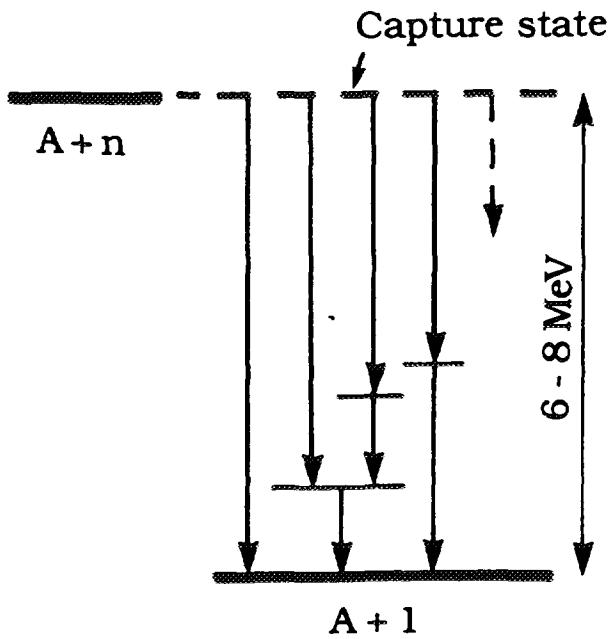

Fig. 1 : Slow neutron capture. Primary transitions are issued from the capture state.

As shown in Fig. 1, if a nucleus A captures a slow (thermal or cold neutron), it will form a nucleus A+1 in an excited (so-called **capture**) state with an energy of about 6-8 MeV above the ground state (except for hydrogen for which it is only 2.2 MeV). This energy is, of course, the same as would be needed to take out the least bound neutron out of the nucleus A+1. The capture state decays first by the emission of **primary** transitions to low-lying excited states, which in turn decay by one or more transitions, called **secondary** transitions, to the ground state. All these transitions, primary and secondary, are characteristic for the capturing isotope and their observation provides therefore **fingerprints** for assessing the presence of particular elements in the sample. Thus the PGAA method consists in observing the primary and/or secondary γ-ray transitions with

the proper instruments. The transition energies and intensity ratios are used for identifying the isotopes (elements) contained in the sample, the intensities for the determination of isotopic (elemental) abundances.

The method is most useful for assessing the presence of low Z elements, presenting a sizable neutron cross section, and which do not produce measurable radioactive products. The interest for low Z elements is because they are not easily observable by X-ray methods (low cross sections, low transition energies). Among the most important applications is the detection of **hydrogen** and of **boron**. As you know well, hydrogen has a high scattering cross section and hence its presence is unwanted in the scattering targets. Since the method is **non-destructive** and has a good sensitivity (see sect. 4), neutrons scatterers are good "customers" of PGAA facilities. Boron has a large neutron capture cross section and can therefore be detected with a very high sensitivity. The PGAA can be used successfully to determine the amount of H and B in metals: this is interesting because even small amounts of these elements can strongly modify their mechanical properties [1]. Let us also mention the importance of boron in the semiconductor industry. PGAA's find also useful applications in nuclear physics (see e.g. [2]).

2. Detection of gamma-rays

The gamma-rays interact with matter by either the photoelectric, the Compton or the pair effect. In the photo-process, the whole incoming photon energy is deposited into the detector. The Compton effect is an inelastic process. The energy released in the detector depends on the scattering angle. It is negligible close to 0° and maximum at 180°. In the pair process an electron-position pair is created. The kinetic energy of these particles is generally deposited in the detector. As the positron reaches a low energy, it annihilates with an electron, creating a pair of 511 keV photons (annihilation radiation). Each of these photons can either escape or interact with the detector.

The detectors which will enter in the construction of the PGAA are:

- scintillators
- semiconductors.

Among the mostly used scintillators are NaI(Tl) and BGO. Sodium iodide, activated with thallium has the highest sensitivity, i.e. the average deposited energy needed to produce a low energy photon is the smallest (see table 1). Consequently this scintillator presents better energy resolution. The schematical response to a monoenergetic gamma radiation is shown in

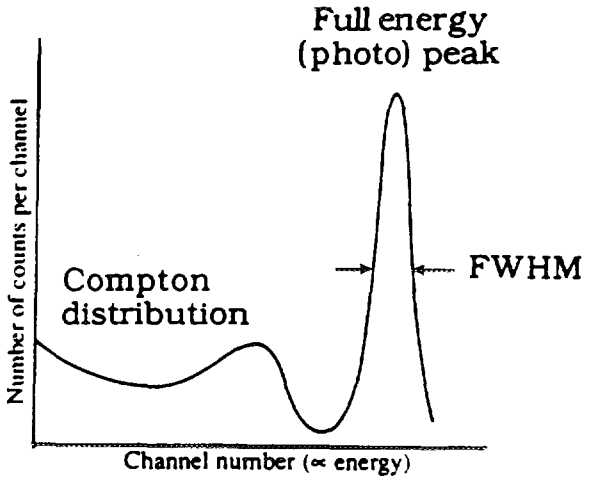

Fig. 2 :
Schematic response of a scintillator to a monoenergetic γ-radiation of energy smaller than the pair-production threshold (1022 keV).

Fig. 2. It is usual to give the resolution for the 662 keV line emitted in the decay of ^{137}Cs. It is of the order of 7 to 12 % for NaI(Tl), depending, in particular, on the detector geometry.

The chemical formula of BGO is $Bi_4Ge_3O_{12}$. Its advantages with respect to NaI(Tl) are its higher density (see table 1) and the high Z elements entering in its composition. The probability of a Compton effect is proportional to the density, favouring a multiprocess leading to a full absorption. The photo-effect cross section is proportional to Z^5 and the pair formation cross section increases with Z^2. The inconvenience, however, is its relatively low sensitivity, i.e. the low energy threshold is as high as about 60 keV. This results also in a poorer energy resolution, about 2 times as large as that obtained with NaI(Tl). Further problems with BGO are that it is very brittle (difficult to machine) and its high cost.

	BGO	CsI(Tl)	NaI(Tl)
Density [g/cm^3]	7.13	4.51	3.67
Linear absorption coefficient [cm^{-1}] at 150 keV	9.98	3.5	2.22
Relat. pulse height (with respect to NaI(Tl)) %	8.10	45	100
Disintegration constant [μs]	0.3	1.0	0.25

***Table 1** : Characteristics of a few widely used γ-ray scintillators.*

Germanium is practically always used in semiconductor γ-ray detectors. Because of its higher density (see table 2) and higher Z it is much superior to silicon, except at very low energies (<50 keV). Because the energy gap is small, Ge detectors have to be used at low temperatures, for practical reasons at liquid N_2 temperature. Germanium detectors have a much better resolution than the best scintillators. It is about 1 keV at ^{137}Cs. It is therefore possible to resolve relatively complicated spectra, as are those observed in on-line spectroscopy.

	Si	Ge
Atomic number	14	32
Density [g/cm^3]	2.42	5.46
Energy gap [eV]	1.10	0.75
Average energy to create an electron-hole pair [eV]	3.6	2.975

Table 2 *: Some characteristics of semiconductor detectors.*

There is, however, one problem: the low energy lines "ride" on top of the Compton distribution produced by the interaction of higher energy transitions in the detector. When the number of lines is large, this Compton background gets large, so that small intensity peaks are not anymore detectable. In addition, the fit of the spectra is then quite difficult because of an

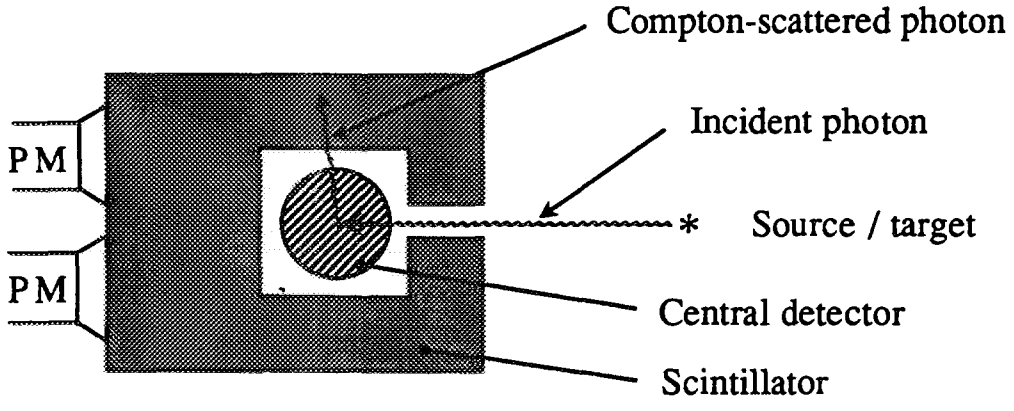

Fig. 3 *: Cut of a Compton-Suppression Spectrometer (CSS) to show its principle of operation.*

undulating background. Fig. 3 shows, in principle, how the background can be reduced by placing the germanium detector inside a scintillator. Suppose that a gamma-ray, emitted by a radioactive source or a target, impinges on the central Ge-detector and produces a Compton interaction. A scattered photon will in general escape (the probability depends on the size of the Ge detector). If it is detected by the scintillator, an electronic circuit will reject the electrical pulse produced by the central detector. Photoelectric events are not rejected and the peak-to-background ratio can be improved up to a factor 10.

Fig. 4 presents part of the γ-ray spectrum emitted following neutron capture in ^{159}Tb observed with a CSS. Without the Compton rejection the background would be much larger, especially at low energy. It must be noted that the spectrum in Fig. 4 presents results from neutron capture in a mono-isotopic target. When several elements (isotopes) are present, the spectra become more complicated. Isotopes with small capture cross section are then difficult to detect and the analysis may involve only the identification of the strongest lines.

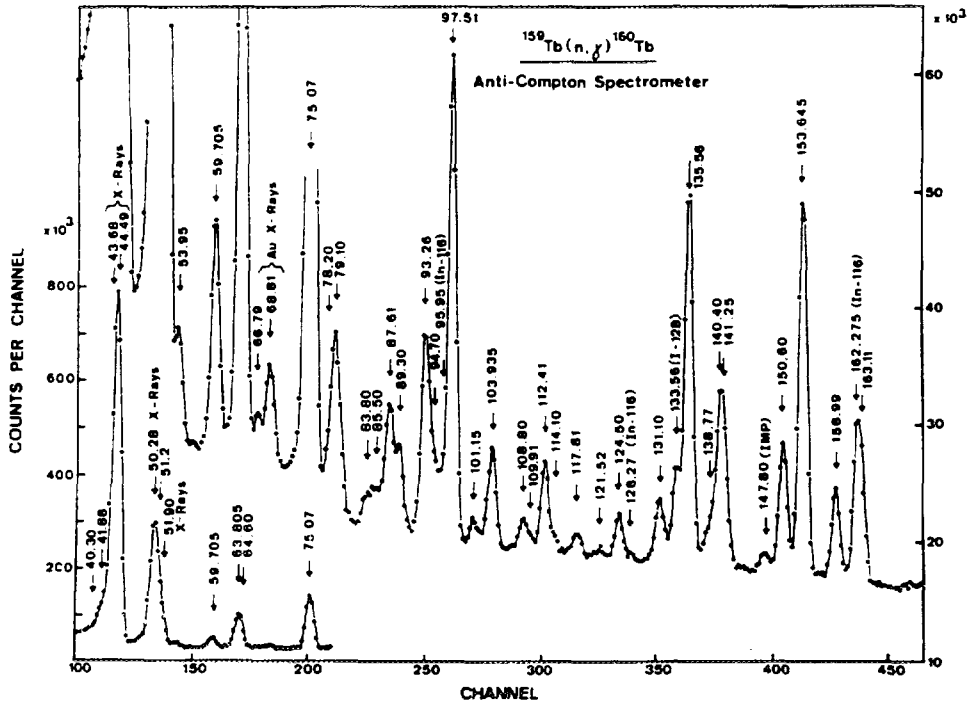

Fig. 4 *: Portion of the low-energy part of ^{159}Tb(n,γ) spectrum observed with a Compton-suppression spectrometer [3]. The final nucleus being doubly odd, heavy and deformed, the spectrum is particularly dense.*

When high-energy ($\gtrsim$ 2 MeV) γ-rays impinge on a detector, the most probable interaction is the pair formation. The full photon energy can be released in the detector if the annihilation radiation is reabsorbed. If only one of the annihilation photon is reabsorbed, a "single-escape peak" will appear 511 keV lower in energy than the full energy peak. If both annihilation photons escape, a "double-escape peak" will appear in the spectrum 1022 keV below the full energy peak (Fig. 5). A monoenergetic transition will therefore produce 3 peaks in the spectrum, in addition to a Compton continuum. This leads to a complicated situation while

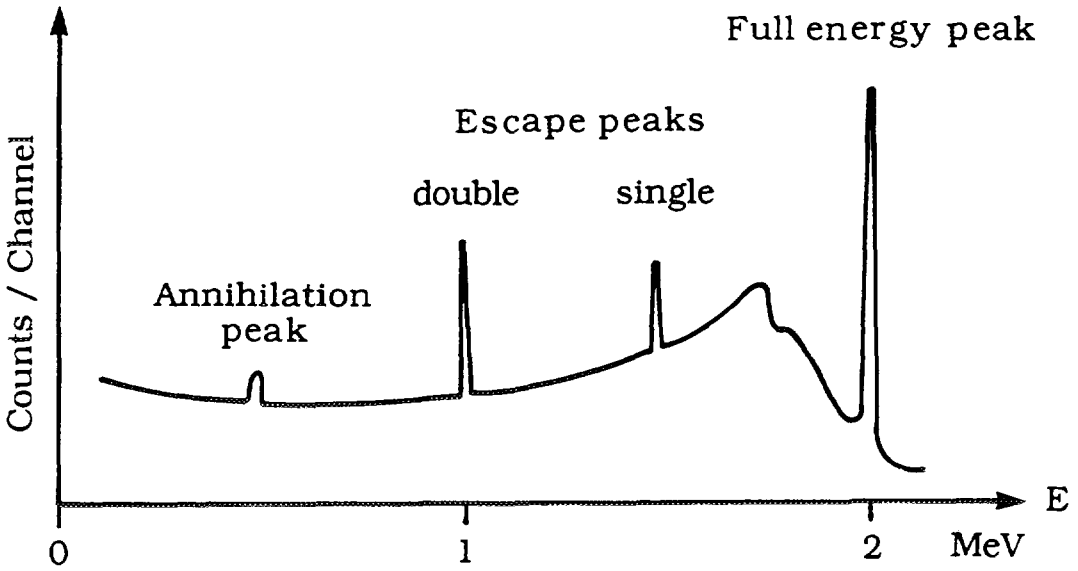

Fig. 5 : Response of a semiconductor detector to a monoenergetic radiation of energy Eγ = 2 MeV. The (broader) annihilation peak is due to interaction of the radiation with matter in the neighbourhood of the detector. The relative intensities of the full energy and of the escape peaks depend on the detector volume and geometry.

observing simultaneously a larger number of transitions. To alleviate this situation a **pair spectrometer** has been devised to select the double-escape peak. The principle is shown in Fig. 6.

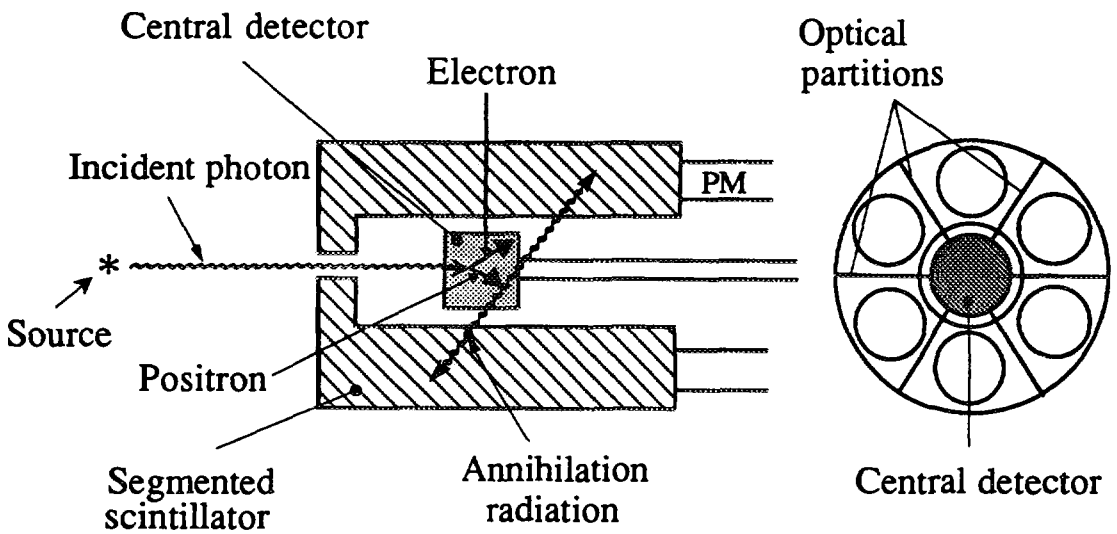

Fig. 6 : Principle of the pair spectrometer.

The germanium detector is placed in the axis of a large, cylindrical scintillator divided optically in six slices, each viewed by a photomultiplier (PM). The selection of a pair event is

performed by requiring a triple coincidence condition between the central detector signal and the 511 keV signals from two opposite slices of the scintillator. This system is very effective.

More detailed information on radiation detectors can be found in refs. [4-6].

3. The PGAA project at SINQ

A horizontal cut of the PGAA project is shown in Fig. 7. The facility will be placed at the end of a half guide. The cold neutron will be focused by a **neutron lens** on a target viewed by a pair and a Compton-suppression spectrometer. The scintillator of the pair spectrometer is made of NaI(Tl) which provides a relatively good selection of the 511 keV annihilation photons. The CSS scintillator will have a part in NaI(Tl) in the rear direction (towards the target) and a part of BGO in the front direction (close to the PM), because the Compton radiation is hard around the 0° (front) and soft in the rear direction (180°).

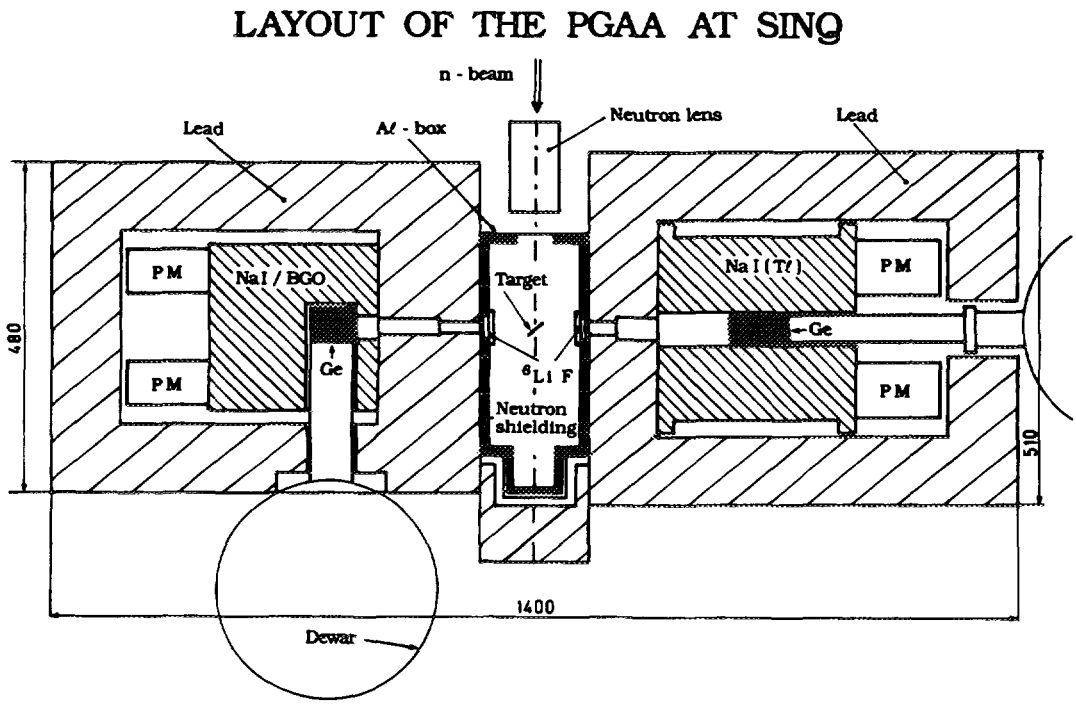

Fig. 7: Schematic plan of the PGAA facility at SINQ

One of the most interesting features is the neutron lens [7]. The principle is shown in Fig. 8. The neutrons are lead through a large number of capillaries just as light in optical fibers. The

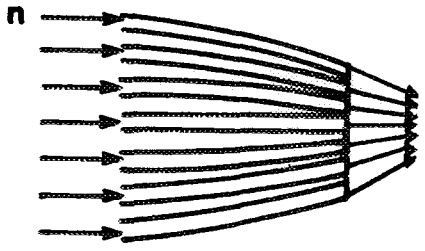

Fig. 8:
Principle of the neutron lens.

capillaries are converging toward a focus. The lens at PSI will produce a neutron spot of less than 1 mm diameter. The flux enhancement will be larger than 30. The use of the lens will provide the following advantages with respect to existing facilities (see e.g. [8,9]):

- increased sensitivity; very small sample of rare material can be analysed
- two-dimensional scanning of samples
- spectroscopy of isotopes with small capture cross sections.

4. Expected performances

Since the PGAA at PSI is not yet operational, we compare in Table 3 a few estimated sensitivity limits with effective numbers measured [2,10] at a facility in Japan [8]. We expect that we will be able to decrease the experimental limits, taking advantage of better conditions and of several experimental improvements.

Element	Predicted [μg]	Observed [μg]	Element	Predicted [μg]	Observed [μg]
H	0.1	1	Mn	0.1	2.2
B	0.0001	0.0023	As	2	9.1
Na	1	4.2	Cd	0.0005	0.007
Al	2	12	In	0.1	0.43
Cl	0.1	0.5	Nd	0.05	0.36
Ti	0.2	0.66	Sm	0.0002	0.003
Va	0.2	1.1	Gd	0.0001	0.0047

***Table 3** : Observed [2,10] detection limits for cold-neutron PGAA compared with predictions.*

Acknowledgments:

The author acknowledges the support of the Swiss National Science Foundation and of the Paul Scherrer Institut.

REFERENCES

[1] D.A. Meyn, Metall. Trans. **5** (1974) 2505.
[2] R.M. Lindstrom et al., Proc. 8th Int. Symp. on Capture Gamma-Ray Spectroscopy and Related Topics, Fribourg 1993, ed. J. Kern (World scientific, Singapore 1994) p. 955.
[3] J. Kern et al., Nucl. Phys. **A221** (1974) 333.
[4] W.R. Leo, Techniques for Nuclear and Particle Physics Experiments (Springer, Berlin Heidelberg 1987).
[5] K. Debertin and R.G. Helmer, Gamma- and X-Ray Spectrometry with Semiconductor Detectors (North Holland, Amsterdam 1988).
[6] C.F.G. Delaney and E.C. Finch, Radiation Detectors (Clarendon Press, Oxford 1992).
[7] M.A. Kumakhov and V.A. Sharov, nature **357** (1992) 390; H. Chen et al. Nature **357** (1992) 391.
[8] C. Yonezawa et al., Nucl. Instr. Meth. **A329** (1993) 207.
[9] G. Molnár et al., J. Radioanal. Nucl. Chem. **167** (1993) 133.
[10] C. Yonezawa, Anal. Sci. **9** (1993) 185.