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Low level γ-ray germanium-spectrometer to measure very low primordial radionuclide concentrations

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Abstract

A new germanium spectrometer especially suited for large sample measurements is described in detail. It is operated in the Gran Sasso underground laboratory under shielding rock of 3300 m water equivalent, which reduces the muon flux by six orders of magnitude. The integral background counting rate in the energy range from 50 to 2750 keV is about 0.15 min^{-1}. The low peak count rates of mostly less than 1 count per day together with a relative efficiency of 102% and the high sample capacity makes this spectrometer one of the most sensitive worldwide. Some sample measurements for the solar neutrino experiment BOREXINO and the detector efficiency calibration by the Monte Carlo method are discussed as well.

Keywords: Low level γ-ray spectroscopy

1. Introduction

Several efforts have been made in the last few decades to reduce the background counting rate of low level germanium spectrometers (Garcia-Leon and Garcia-Tenorio, 1994; Heusser, 1994). Careful material selection combined with a close cooperation with the manufacturer and going underground are the most important steps. An additional prerequisite is the reduction of ^{222}Rn, which can diffuse through the standard shield and contributes considerably, via the daughters (^{214}Pb, ^{214}Bi), to the background count rate.

Mainly in the context of the solar neutrino experiment BOREXINO (Arpesella et al., 1991; Benziger et al., 1996), this low level germanium spectrometer has been constructed. The spectrometer is operated in the Laboratori Nazionali del Gran Sasso, an underground laboratory located in the Abruzzese mountains, 150 km north-east of Rome. The shielding by the rock of about 3300 m water equivalent (m w.e.) reduces the muon flux by six orders of magnitude to approximately 1 μ/m^2 h.

BOREXINO is an extreme low level liquid scintillation detector with very low impurities of U and Th (10^{-16} g/g). Its components need severe selection for high radiopurity. High sensitivity γ-ray spectroscopy is an ideal tool for that purpose.

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2. Detector system

The design of the spectrometer was choosen to fulfill the following criteria:

— extreme low intrinsic contamination,
— minimize cosmogenic production in detector and shielding copper,
— strong reduction in the external background with special attention to radon supression,
— high specific sensitivity by large sample capacity.

The p-type high purity germanium crystal of the spectrometer has a mass of 2.2 kg (corresponding to a volume of 0.7 l) with a certified relative efficiency of 102% (Canberra Semiconductors N.V., Olen, Belgium). The detector construction was performed in close cooperation with Canberra. Careful material selection was carried out at the low level laboratory of the MPI Heidelberg (shielded by 15 m w.e. overburden). The cooling rod forms a right angle below the crystal to avoid the direct line of sight to the outside including the preamplifier. The latter is even outside the whole shield because of its less radiopure components. The copper and lead used were stored underground (in Heidelberg) and taken out only for purification and fabrication.

The sample chamber is formed by plates of electropolished copper with a thickness of 5 cm (base plate: 7 cm). The inner dimensions of 25 cm × 25 cm × 33 cm allow one to place about 13 l of sample around the crystal housing. Four layers of low-activity lead, each 5 cm thick, are placed around the copper. The innermost layer and the central parts of the second layer consist of lead from the company Plombum (Krakov, Poland) with a ^{210}Pb contamination of 5 Bq/kg. The other layers are made of lead from Boliden, Graviner and D'Overpelt with 23–32, 23 and 133 Bq/kg of ^{210}Pb (Kolb, 1987). The sample chamber is closed on top with two large copper plates carrying the lead. They can be slided aside on a steel frame by the aid of slide bearings to open and close the chamber. Fig. 1 shows a cut view through the lower part of the spectrometer. The total mass of copper and lead is about 0.5 and 4.6 t, respectively.

The arrangement is housed in an airtight steel box consisting of three parts: a box of 1 m ×1 m ×1.2 m around the lead/copper shield, an operating box on top of the latter and an airlock for access to the samples. The whole system is flushed permanently by gaseous nitrogen from standard 50 l gas cylinders starting inside the measuring chamber to suppress radon influx from small air leaks. The airlock is separately flushable with nitrogen.

Inside the airlock and above the sample chamber are moveable tables for easier handling of large and heavy samples. A crane is mounted on top of the operating box to handle loads up to 50 kg. It can be used to lower heavy samples into the chamber. The samples are first stored above the closed sample chamber in

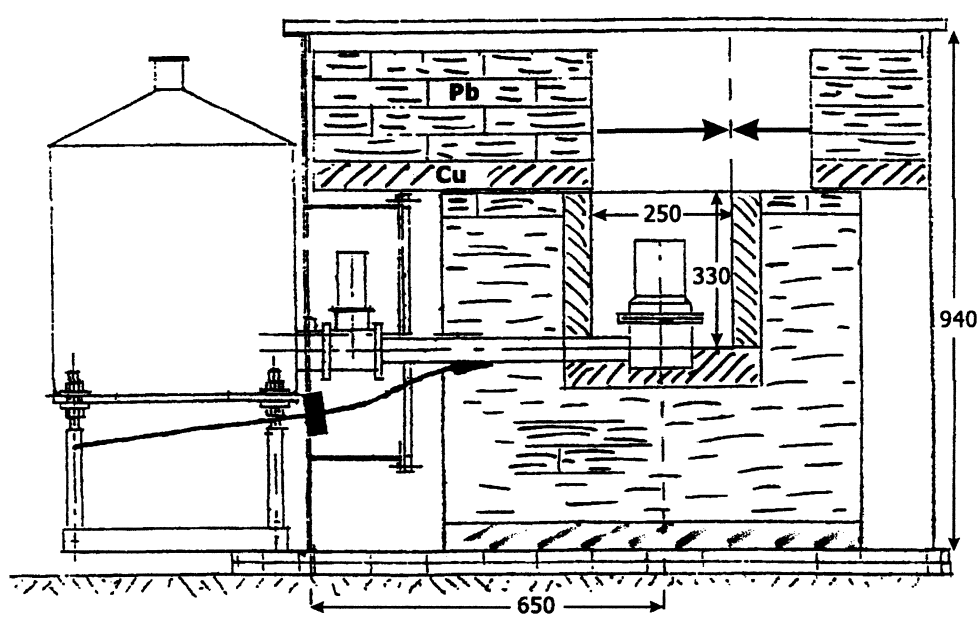

Fig. 1. Cut view of the spectrometer. Arrows indicate the position of the moveable upper shielding door when the chamber is closed. Dimensions are given in mm.

nitrogen, so that the plate out Rn progenies or trapped radon can decay before the sample is measured. Windows and three glove-systems complete the operating box, see Fig. 2.

A calibration source can be fed into the inner chamber in a guiding Teflon tube. The source is fixed at the end of a steel wire and can be moved remotely from outside the shield, directly below the detector, by means of a motor.

3. Measurements

3.1. Background measurements

Fig. 3 shows some background spectra measured at the low level laboratories (LLL) of Heidelberg (upper two spectra) and at Laboratori Nazionali del Gran Sasso (GS). From top to bottom are displayed: a spectrum of the detector without shielding, with a provisional shield (15–20 cm lead) and a recent spectrum with complete shielding and nitrogen flushing. Note the logarithmic scale and the difference of about five orders of magnitude between the uppermost and the lower spectrum.

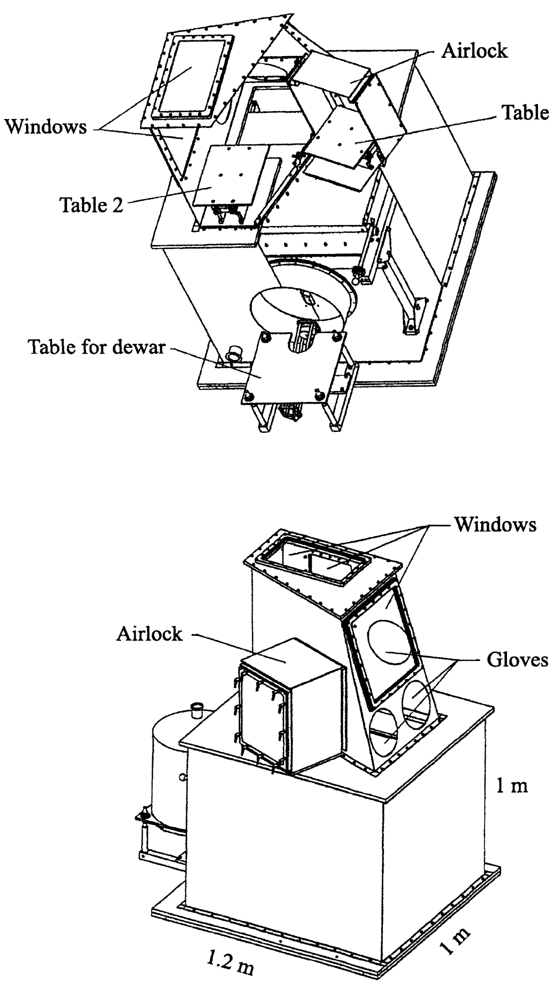

Fig. 2. View of the spectrometer enclosure from the top (partly cut open) and from the side, see text for details.

Table 1
Integral background counting rates (50–2750 keV) for several measuring periods and configurations. The labels correspond to the ones given in Table 2. Measurement 'C' was taken with Ge-bricks inside the chamber, 'A', 'B' without. Uncertainties correspond to 90% confidence level

Label	Measuring period	Lifetime (days)	Integral counting rate (min^{-1})
A	Autumn 1997	51.16	0.179 ± 0.003
B	Spring 1999	34.36	0.150 ± 0.003
C	November 1998	11.92	0.093 ± 0.004

The integral counting rate at Gran Sasso has decreased from an initial value of 0.179 min^{-1} (50–2750 keV) to 0.150 min^{-1}, due to the decay of short living cosmogenic nuclei, see Table 1 for further information. The counting rates for single lines of several nuclides are given in Table 2. All counting rates of the 'actual' spectrum ('B') are below 1 per day, except for the ^{226}Ra progenies. The 811 keV line of the cosmogenic ^{58}Co ($T_{1/2} = 70.6$ days) has decreased completely, whereas ^{60}Co ($T_{1/2} = 5.27$ years) is still present.

To investigate if the residual lines of the ^{226}Ra progenies are due to insufficient flushing, Rn contami-

Table 2
Counting rates for single lines of different nuclides. The labels correspond to the measurements given in Table 1, uncertainties and upper limits are given with 90% confidence level

Isotope	E_γ (keV)	Counting rate (per day)		
		A	B	C
^{238}U-chain				
^{226}Ra	186	< 0.45	< 0.55	< 0.9
^{214}Pb	352	1.19 ± 0.49	0.91 ± 0.60	< 0.5
^{214}Bi	609	1.70 ± 0.40	1.62 ± 0.54	0.50 ± 0.45
^{232}Th-chain				
^{228}Ra	911	< 0.22	< 0.23	< 0.2
^{212}Ra	727	0.22 ± 0.20	< 0.21	< 0.4
^{208}Tl	583	< 0.29	< 0.23	< 0.3
^{58}Co	811	2.24 ± 0.40	< 0.19	0.3 ± 0.3
^{60}Co	1173	0.78 ± 0.26	0.54 ± 0.26	0.6 ± 0.4
	1333	0.66 ± 0.25	0.76 ± 0.33	0.4 ± 0.3
^{40}K	1461	0.76 ± 0.22	0.56 ± 0.23	0.6 ± 0.4

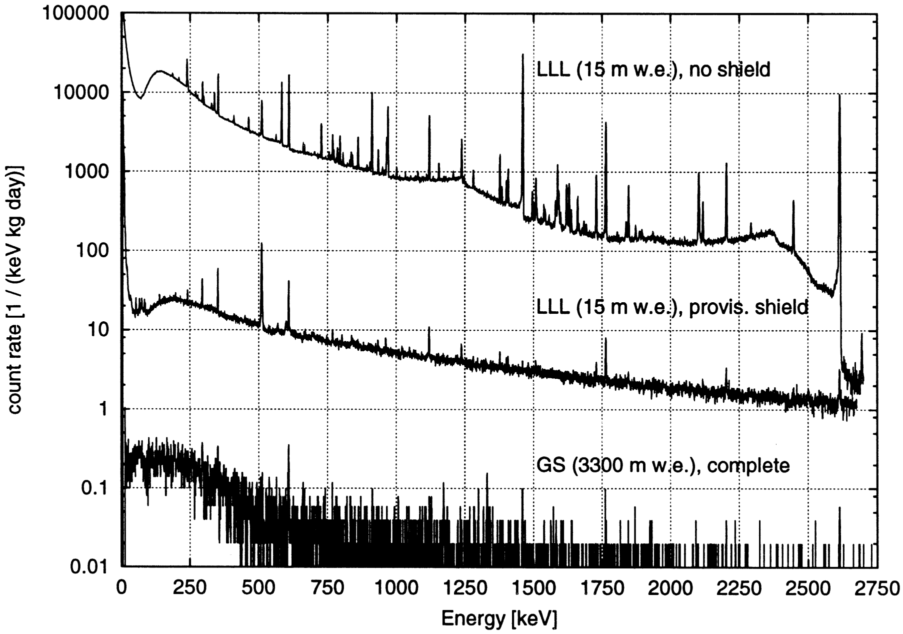

Fig. 3. Background spectra measured at different locations and different shielding configurations, see text for details.

nation in the nitrogen or originating from surface contamination of the copper, the chamber was filled (almost completely) with bricks made from high purity germanium. The integral count rate decreased significantly (see Table 1), but since the statistics of the Rn progenies lines are as yet insufficient to draw a clear conclusion, the measurements have to be continued. Further investigations by replacing the nitrogen[1] with high purity nitrogen is planned.

3.2. Sample measurements

Out of the many samples measured for BOREXINO two examples are given in Table 3, which are representative for the performance of the spectrometer. The first sample, a stainless steel disk of 8 cm diameter, 0.6 cm height and 0.237 kg mass was measured lying flat on top of the detector. The second sample, a large stainless steel foil, rolled into a cylinder with inner and outer diameters of 12.2 and 21.2 cm and height 23.3 cm (38.1 kg mass), was placed around the detector.

The efficiency for the different isotopes was calculated with a Monte Carlo code (see next section). The spectra were analyzed with a program dedicated to low level spectra (Neder, 1998). Some of the results, together with corresponding efficiencies, are shown in Table 3. The efficiencies were calculated including the decay schemes, so that coincidence summing is included. The effect of the latter on different geometries can be seen by comparing the 609 and the 661 keV line (^{137}Cs emits only a single gamma).

Although the efficiency for the steel foil is much lower, the specific sensitivity is higher due to the larger mass. The large uncertainties in the specific activity reflects the low statistics, for example the 609 keV line of ^{214}Pb for the steel foil coil had a count rate of only (3.0 ± 2.5) per day.

4. Simulations

To determine the efficiencies for the different sample geometries we use Monte Carlo simulations based on the well known code GEANT (CERN, 1995). This method has the advantage that laborious preparation and measurement of a calibration source for each new sample configuration can be avoided.[2]

For that purpose we have implemented the complete spectrometer and some standard sample geometries in the code. A new geometry (including material and density information) can be added very easily. The isotopes are simulated including their decay schemes[3]

[1] Nitrogen from standard bottles can have a ^{222}Rn contamination of up to 1 mBq/m^3 (Freudiger, 1998).

[2] And it is even faster. A complete analysis needs only a few hours.

[3] At least all gammas with emission probabilities greater or equal 1% are implemented.

Table 3
Selected efficiencies (normalized to 100% emission probability) and results (calculated from all detectable limits) for two measured stainless steel samples (see text). The uncertainties correspond to 90% confidence level

	Efficiency (%)			Specific activity (mBq/kg)		
	609 (keV)	661 (keV)	1461 (keV)	^{226}Ra	^{137}Cs	^{40}K
Steel disk	4.9 ± 0.2	6.6 ± 0.1	4.4 ± 0.1	17.0 ± 2.5	0.6 ± 0.6	4 ± 6
Steel foil coil	0.42 ± 0.01	0.46 ± 0.08	0.41 ± 0.02	0.64 ± 0.21	< 0.12	1.8 ± 0.6

(Firestone, 1996) so random coincidence summing effects are already taken into account. Generally we simulate 10^6 decays for each interesting isotope, which takes about 10–30 min CPU-time on a DEC α workstation.

Comparisons with calibration samples (voluminous and point sources on top) showed a relative agreement better than 5% (Neder, 1998). Further investigations and checks are in progress, e.g. an inclusion of angle correlation and a better 'fit' of the detector parameters (thickness of dead-layer and so on).

5. Conclusions

The described low level germanium spectrometer has the capability to measure extraordinarily large (13 l) and heavy samples (50 kg) at an extremely low background. The latter has been achieved by careful material selection and effective radon suppression. The counting rates of single background lines are below 1 per day except for the ^{222}Rn progenies. Sample measurements showed that a specific sensitivity of less than 1 mBq/kg can be reached.

The combination of high sample mass and low background makes this spectrometer an ideal tool for material selection for ultra-low background experiments as e.g. BOREXINO.

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