Highly sensitive bolometers for rare alpha decay studies

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Abstract. High resolution detectors able to identify background events are very appealing in the study of rare nuclear processes. Scintillating bolometers featuring simultaneous read-out of heat and scintillation signals, can effectively address this problem thanks to the possibility to discriminate different ionizing particles and achieve background free experiments. With this technique it has already been possible to measure rare alpha decays never observed before or improve by orders of magnitude the existing limits.

1 Scintillating bolometers for rare phenomena studies

The detection of rare $\alpha$ decays with half-lives $>10^{19-10^{20}}$ y represents a big challenge from the experimental point of view. Indeed, standard detectors such as gas counters, scintillation detectors or semiconductor devices have difficulties in achieving the required sensitivity for this kind of study because of the background.

The main sources of background can be divided into two big classes: cosmic rays and environmental radioactivity. The former is composed mainly by muons and the installation of the detectors in deep underground laboratories it is usually enough to greatly reduce this source of background to a negligible level. For example the average 1400 m rock coverage of the Laboratori Nazionali del Gran Sasso (LNGS) gives a reduction factor of $\sim10^6$ in the cosmic ray flux. The muons residual flux is $\sim1 \mu/m^2/h$. For what concern the environmental background the main sources are $\beta$ and $\gamma$ events due to natural radioactivity. These are particularly troublesome especially in the study of low energy $\alpha$ decays with Q value lower than the highest gamma line due to natural radioactivity (the 2615 keV line of $^{208}$Tl). The shielding that could be provided (e.g. by high-Z material such as lead and copper) is not enough to reach extremely high sensitivity. In addition, the construction materials of the detector assembly and the shielding itself must be considered. In spite of a careful selection of these materials, its radioactive contamination cannot be neglected. Finally, it has to be mentioned that the neutrons induced background is very low in the region of interest and, if necessary, can be easily made negligible through appropriate neutron shielding.

In the case of scintillation detectors the situation is even worse because of the lower light yield of $\alpha$ particles that makes the position of the searched peak further down in the energy spectrum where the background is usually higher.

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Moreover, the sensitivity of detectors in which the radioactive source is external to the detector itself, as for example Si surface detectors, is limited by the low detector efficiency and by the low energy resolution due to the energy lost by the particle before hitting the detector.

All these limitations can be easily overcome by using scintillating bolometers. They have several advantages: the crystal growth can be optimized to have large and radio-pure detectors, which means large source mass and low intrinsic background. In addition, bolometers can be grown, in principle, with any interesting isotopes. This allows to study $\alpha$ decays in bolometers with mass of hundreds of grams with high energy resolution and with a detection efficiency $\sim 1$. Finally, the simultaneous readout of light and heat signals results in a powerful tool for background identification.

Scintillating bolometers can be sketched as a calorimetric absorber based on the isotope of interest and a light detector able to measure the emitted photons. The driving idea of this hybrid detector is to combine the two information available: the heat (i.e. the large fraction of energy converted into phonons) and the emitted scintillation light (i.e. that small fraction of the energy which is converted into photons). Thanks to the different scintillation yield of different particles ($\beta/\gamma/\mu$, $\alpha$ and neutron) they can be very efficiently discriminated.

The usual way to present the results obtained with scintillating bolometers is to draw the heat vs. light scatter plot (figure 1). Each event is identified by a point with abscissa equal to the heat signal (recorded by the main bolometer), and ordinate equal to the light signal (at the same time recorded by the light detector).

![Figure 1](image.png)

Figure 1. Typical heat vs light scatter plot obtained with scintillating bolometers with mass of hundreds of grams. In the scatter plot it is possible to identify clearly the alpha peaks and the beta/gamma region characterized by the gamma peaks and by a continuum due to beta events and gamma events that don’t release all their energy in the crystal (e.g. Compton events).

1.1 Experimental setup

The results reported in the following on $^{209}$Bi excited state and lead isotopes were obtained by using similar experimental setups. Crystals were held by means of PTFE supports to a Cu structure and were surrounded (with no direct contact) by a plastic reflecting foil. The light detector was a thin pure Ge crystal absorber working as a bolometer [1]. The temperature sensor for both the main crystal and the light detector were Neutron Transmutation Doped (NTD) germanium thermistor.

The crystals were run at $\sim 10$ mK in an Oxford $^{3}$He/$^{4}$He dilution cryostat deep underground in the LNGS. The external shield consisted of $10$ cm of lead surrounded by a neutron shield of $\sim 7$ cm of polyethylene and about $2$ cm of CB$_4$. A $\sim 5.5$ cm Roman lead shielding was placed inside the cryostat just above detectors, $\sim 1.2$ cm on the sides and $\sim 3$ cm just below in order to shield the bolometers from $\gamma$ radiation due to contaminations of the cryostat materials.

The amplitude and the pulse shape of the signals were determined by the off-line analysis. To maximize the signal-to-noise ratio, the pulse amplitude was estimated by means of the optimum filter technique.

The energy spectrum of the main crystal was calibrated attributing to each identified peak the nominal energy of the $\gamma$ lines. The calibration of the light detector was obtained by means of a weak $^{55}$Fe source placed close to the Ge wafer that illuminated homogeneously the face.
This experimental setup was used to study many scintillating bolometers for neutrinoless double beta decay such as ZnMoO$_4$ [2], ZnSe [3], CdWO$_4$ [4] and CaMoO$_4$ [5]. This setup was also used to study materials surface contaminations with a Bi$_4$Ge$_3$O$_{12}$ (BGO) crystal [6]. In a long background measurement performed with this last compound the $\alpha$ decay of $^{209}$Bi on the excited state of $^{205}$Tl was observed for the first time [7].

2 Rare $\alpha$ decay of $^{209}$Bi

$^{209}$Bi was thought to be the heaviest stable isotope, until the first evidence of its decay was obtained by means of a BGO scintillating crystal [8]. This measurement provided the half-life of the ground state (GS) decay. However, $^{209}$Bi is expected to decay also with a transition to the 204 keV excited level of $^{205}$Tl (ES). The simultaneous observation of the two decays was made possible thanks to an 889.09 g BGO crystal operated as scintillating bolometer in the LNGS. The crystal was faced to a high purity Ge slab (36 mm diameter and 0.3 mm thickness) used as light detector. The composite device was characterized by an energy resolution of about 37 keV for the BGO and 0.5 keV for the light detector, a high detection efficiency and a large light output for $\gamma$ rays (16.61 ± 0.02 keV/MeV). These excellent performances allowed to distinguish between the ground and the excited state transitions.

About 375 hours of background measurement were collected. Several very intense lines are visible in the $\gamma$ spectrum (and were used for the heat spectrum energy calibration). They are due to the internal contamination of the crystal in $^{207}$Bi (produced by cosmic ray protons interaction on $^{206}$Pb [9]) and the background $^{40}$K and $^{232}$Th lines.

The structure of the $\alpha$ region appears slightly more complicated than the $\beta/\gamma$ one. Here we can identify two different kind of events.

Pure $\alpha$-decays are aligned along the same curve in the heat vs light scatter plot. $\alpha$-decays in crystal bulk are monochromatic with an energy corresponding to the Q-value of the decay (since both the energies of the emitted alpha and of the recoiling nucleus are detected). Two such lines are clearly evident in the scatter plot reported in [7] and are identified as due to $^{209}$Bi $\rightarrow$ $^{205}$Tl decay and to $^{210}$Po $\rightarrow$ $^{206}$Pb decay which is present in the crystal probably as a result of $^{209}$Bi activation [10].

For $\alpha$-decays on the excited state in which the $\gamma$ is fully absorbed, the light signal is higher than for a pure $\alpha$ emission because of the higher light yield of the $\gamma$. An example is the $^{210m}$Bi decay. The isotope $\alpha$-decays to different excited levels of the daughter isotope ($^{206}$Tl) with the contemporary emission of one or more $\gamma$ ray.

Similar to $^{210m}$Bi is the case of $^{209}$Bi decay. It follows two different paths to the $^{205}$Tl ground state. Ground state decay produces an $\alpha$ particle plus a recoil (Q=3137 keV); being a monochromatic pure $\alpha$-decay it produces a line in the $\alpha$-band (the probability of fully contain both the $\alpha$ particle and the recoiling nucleus inside the crystal is obviously $\sim$1). $\alpha$-decay on the excited state produces an $\alpha$ particle plus recoil (Q=2933 keV) and a prompt $\gamma$ ray ($E_\gamma$=204 keV). In (92.1±0.5)% of cases the $\gamma$ photon is fully absorbed in the BGO crystal.

The branching ratio for GS-GS transition results to be (98.8±0.3)%. Taking into accounts for both the trigger efficiency and the pulse-shape cuts efficiency, the detection efficiency was (87±2)%. The half-life for the GS-GS transition results $T_{1/2}^{GS-GS}$=(2.04±0.08)$\times$10$^{19}$ years in good agreement with the previously reported one [8].

3 Lead isotopes

As a result of the observation of $^{209}$Bi $\alpha$ decay, lead is considered to be the heaviest stable element. However $\alpha$ decay in lead is energetically allowed for all the four naturally occurring isotopes ($^{204}$Pb,
$^{206}\text{Pb}$, $^{207}\text{Pb}$ and $^{208}\text{Pb}$). The theoretical expected half-lives are $>10^{35}$ y and therefore there is no feasible perspective to observe the $\alpha$ decay of these isotopes. However, a measurement performed with a PbWO$_4$ crystal allowed to improve considerably the half-life limits and to test the reliability of the various nuclear models.

The PbWO$_4$ crystal measured in the LNGS was 3.0×3.0×6.1 cm$^3$ for a total mass of 454.1 g [11]. A tiny splint ($\sim$ 50 mg) was removed from the crystal and analyzed through ICP-MS in order to evaluate the isotopic abundances of the four lead isotopes. This crystal was grown with ancient Roman lead [12] because of the low activity in $^{210}\text{Pb}$. The radioactivity of commercial lead can be of the order of few tens of Bq/kg for ore-selected samples while, since the $^{210}\text{Pb}$ half-life is 22.3 y, its activity is extremely small in ancient samples. The $^{210}\text{Pb}$ content of this Roman lead was measured to be less than 4 mBq/kg. A high activity in $^{210}\text{Pb}$ is indeed a limiting factor in bolometric measurements because of the relatively slow time response of these devices which is of the order of hundreds of ms. A ‘high’ rate with a slow time response induces troublesome pile-up effects.

The total live time of the background measurement was 586 hours. No $\alpha$ peaks that could be ascribed to lead isotopes decay was observed. Therefore new more stringent upper limits on the half-lives of four natural lead isotopes were estimated at 90% C.L. with the following sensitivities:

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T_{1/2}(^{204}\text{Pb}) > 1.4 \cdot 10^{20} \text{y}
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$$
T_{1/2}(^{207}\text{Pb}) > 1.9 \cdot 10^{21} \text{y}
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$$
T_{1/2}(^{206}\text{Pb}) > 2.5 \cdot 10^{21} \text{y}
$$
$$
T_{1/2}(^{208}\text{Pb}) > 2.6 \cdot 10^{21} \text{y}
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4 Conclusion

Recently, measurements of $\alpha$ decays with half-lives $>10^{19}$-$10^{20}$ y demonstrated the high sensitivity achievable with scintillating bolometers for the discovery of rare nuclear processes. The main advantage of this technology is the wide choice of detector materials that allows to investigate with an homogeneous technique isotopes that are not easily measurable with conventional detectors. Moreover, the simultaneous readout of light and heat signals results in a powerful tool for background identification. This is fundamental in case the expected energy of an $\alpha$ decay lies in the environmental $\beta/\gamma$ energy region. As examples, the measurement of the half-life of the $^{209}\text{Bi}$ and the limits on lead isotope half-lives were reported.

References