First Measurement of the Partial Widths of $^{209}$Bi Decay to the Ground and to the First Excited States

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$^{209}$Bi alpha decay to the ground and to the first excited state have been recently observed for the first time with a large BGO scintillating bolometer. The half-life of $^{209}$Bi is determined to be $\tau_{1/2} = (2.01 \pm 0.08) \times 10^{19}$ yr while the branching ratio for the ground-state to ground-state transition is $(98.8 \pm 0.3)$%.

I. Introduction.—$^{209}$Bi is the only naturally abundant isotope of bismuth. Its $\alpha$ decay to $^{205}$Tl was predicted on the basis of the $^{209}$Bi mass excess, but escaped direct observation until 2003 when it was observed with a small BGO scintillating bolometer; Ref. [1]. In that work—due to the small mass of the detector—only the partial width for the ground-state decay was measured, resulting in a half-life of $(1.9 \pm 0.2) \times 10^{19}$ yr. The decay was in that case ascribed to $^{209}$Bi on the basis of the energy measured for the emitted $\alpha$ line; interferences from other $\alpha$-decaying isotopes—like $^{190}$Pt and $^{186}$Os—were excluded making reasonable assumptions on the purity of the crystal.

However, the decay can proceed either as a direct transition to the $^{205}$Tl ground state (GS-GS transition) or as a transition to the first excited level of $^{205}$Tl (GS-ES transition); see Fig. 1. The contemporary observation of both the lines would provide—as observed also in Ref. [1]—the real conclusive test on the identification of $^{209}$Bi decay. In this Letter we report the first experimental evidence for $^{209}$Bi decay to the 204 keV excited level of $^{205}$Tl, the measurement of the branching ratio for the two transitions, and finally the evaluation of $^{209}$Bi half-life. This result, besides providing a further compelling evidence on the existence of $^{209}$Bi decay, adds a new experimental input for the study of the alpha decay of odd-odd nuclei where a non-negligible angular momentum is carried by the emitted alpha particle.

II. Experimental technique.—The detector used for this study is a BGO scintillating bolometer. This is realized instrumenting a Bi$_4$Ge$_3$O$_{12}$ crystal with a temperature sensor and coupling it to a light detector (LD). Both the BGO crystal and the LD operate as bolometers, at a temperature of few tens of mK. The working principle is quite simple: the energy released by an ionizing particle that traverses the BGO crystal is converted both into scintillation light and into heat. The former gives rise to a light pulse that is recorded by the LD, the latter produces a temporary temperature increase of the BGO crystal that is recorded by its temperature sensor. The fraction of the total deposited energy spent in scintillation depends on the nature of the particle and is called light yield (LY), $\beta$’s and $\gamma$’s have the same LY, which is typically different from that of $\alpha$’s or neutrons; in this way heat and light signals can be used to disentangle particle identity. At room temperature BGO crystals produce about $(8–10) \times 10^3$ scintillation photons for a 1 MeV electron, corresponding to a LY of the order of 20–26 keV/MeV. The scintillation yield increases when cooling the crystal [2].

A. Detectors and experimental setup: The design of the single BGO bolometer and of the LD follows that of the other macrobolometers operated in the past by our groups (Refs. [3,4]). The main bolometer is a $5 \times 5 \times 5$ cm$^3$ BGO
crystal (with a mass of 889.09 g) purchased from SICCAS (China) with optical grade surfaces. The crystal is secured inside a copper holder and is surrounded on 5 faces by a light reflector (3M VM2002). Attached to the crystal is an NTD Ge thermistor (Ref. [5]) that acts as a thermometer: the temporary temperature increase of the crystal—following particle interaction—produces a voltage pulse with an amplitude proportional to the deposited energy. The LD is a bolometer built with the same technique and optimized in order to be able to detect the small energy carried by the scintillation photons. The LD consists in a high purity Ge wafer, 36 mm diameter and 0.3 mm in thickness. On the side facing the scintillating crystal the surface of the wafer is “darkened” with a deposition of a 600 Å layer of SiO₂ to improve light absorption. An NTD Ge thermistor for the temperature signal readout is glued on the other side of the LD, to which a ⁵⁵Fe source for energy calibration is faced.

The BGO and the LD bolometers (namely, the heat and light channels) are provided with two completely independent readout chains. The voltage signal produced on the NTD thermistors are amplified and filtered by the front-end electronics and fed into the analog-to-digital converter. When the trigger fires, the entire signal waveform is sampled, digitized, and saved to a disk. Pulse-height and pulse-shape parameters are computed by the off-line analysis for each acquired event. Signals deformed by excess noise or by pileup are rejected by the analysis since they can produce a broadening or a deformation of the peaks, spoiling the energy resolution. The true rate is then reconstructed measuring the trigger efficiency and the probability that a pulse-shape cut rejects a true particle event. The former is measured on pulser generated events, the latter is obtained by comparing the event rate in a background peak before and after pulse-shape cuts (the ratio between the two rates is the pulse-shape cut efficiency).

B. Energy calibration: The energy content of the heat and light signals are evaluated using calibration data.

The light signal is calibrated using ⁵⁵Fe x rays, assuming a linear dependence of the pulse height on energy. This is an approximation since in general the relationship between energy and pulse height is not linear (the exponential dependence of the thermistor resistance on temperature being the main source of nonlinearity). The calibration returns the energy content of the recorded scintillation signal. It should be noted that the energy is not corrected for the light collection efficiency; therefore, it does not provide an absolute determination of the amount of scintillation light produced by the BGO crystal.

The heat signal is calibrated using the more intense γ lines visible in the BGO heat spectrum. They cover a range from 0.5 to 2.6 MeV and are produced by radioactive contamination of the crystal or of the cryogenic setup. Above and below the calibration is obtained by extrapolation. It is important to remark that in calibrating the heat signal the nominal γ line energy is assumed for each full energy peak observed in the heat spectrum. This means that the calibration—for β/γ events—returns the total amount of energy released by the interacting particle in the crystal (although this energy is only partially converted into heat). On the contrary for α events it overestimates the true total energy, since the LY of α particles (LYα) is different (lower) than for β/γ (LYγ) (for a detailed discussion of this issue see [3]). To emphasize this fact, the heat axis units are indicated as keVβ/γ.

III. Results.—The detector was operated in the cryogenic facility installed in Hall C of Laboratori Nazionali del Gran Sasso (L’Aquila, Italy). The BGO crystal cooled very slowly (while the cryostat—as usual—reached the base temperature in few days), a behavior that can be ascribed to some excess heat capacity that decouples at low temperatures.

We collected 374.6 hr of background measurement in semistable conditions: the crystal was at 30 mK, still cooling during the measurement period improving its signal height by ~30%. Off-line corrections—using the technique described in Ref. [6]—were applied to account for the thermal drift observed during the measurement.

The light versus heat scatter plot corresponding to this statistics is shown in Fig. 2. The α and γ regions appear to be clearly separated, their light and heat spectra are reported in Fig. 3.

A. Gamma region: Several very intense lines are visible in the γ spectra (and were used—as discussed above—for the heat spectra energy calibration). They are due to the internal contamination of the crystal in ²⁰⁷Bi and the

FIG. 2 (color online). Light versus heat scatter plot corresponding to 374.6 hr of background (no external sources) measurement. The heat axis is calibrated with gamma lines, as described in the text; the subscript β/γ on the heat “keV” units indicates that the calibration gives the correct energy only for β/γ particles. Colors are used to highlight the α band. Pure α decays lie on a curve that is fitted with a degree two polynomial. Mixed α + γ events lie above this curve; they are highlighted in a lighter color. The inset shows ²⁰⁹Bi decay events.
background, \(^{40}\)K and \(^{232}\)Th lines (usually observed in all measurements and ascribed to detector + cryostat contamination). \(^{207}\)Bi is a common contaminant already observed in BGO crystals which is produced by cosmic ray protons interaction on \(^{206}\)Pb; Refs. [7,8]. The average FWHM energy resolution on the heat channel—as measured on the more prominent \(\gamma\) lines—is \((37.5 \pm 0.5)\) keV, with no evident dependence on energy.

The energy resolution of the LD, measured on the \(^{55}\)Fe line, is 0.5 keV. The FWHM of the scintillation peaks produced by the \(^{207}\)Bi, \(^{40}\)K, and \(^{232}\)Th \(\gamma\) photons interacting in the BGO ranges from 0.8 to 1.5 keV. The LY has no evident dependence on energy; its average value is \(\overline{LY} = (16.61 \pm 0.02)\) keV/MeV. The overbar here indicates that LY is not corrected for the light collection efficiency.

\textbf{B. Alpha region:} 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 (no \(\gamma\) emission) are aligned along the same curve in the scatter plot (see Fig. 2). These events are produced by \(\alpha\) particles impinging on the crystal from an external source, or by \(\alpha\) decays in crystal bulk. The former have generally a continuous energy distribution (see, for example, Ref. [9]); the latter 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 our scatter plot and are identified as due to \(^{209}\)Bi \(\rightarrow\) \(^{205}\)Tl decay \((Q = 3137.2 \pm 0.8\) keV, Ref. [10]), and to \(^{210}\)Po \(\rightarrow\) \(^{206}\)Pb decay \((Q = 5407\) keV) which is present in the crystal, probably as a result of \(^{209}\)Bi activation (see Ref. [11]). Since \(\alpha\) particles have a LY that is lower with respect to the \(\beta/\gamma\) one, the two lines appear in the heat spectrum with energies higher than the nominal ones. We recall that this effect is simply due to the procedure adopted for the heat axis calibration. The LY \(\alpha\) and the corresponding quenching factors (QF = LY\(_{\alpha}\)/LY\(_{\gamma}\)) are

\[
^{209}\text{Bi} \left[ \begin{array}{c} \overline{LY}_{\alpha} = 2.482 \pm 0.002 \text{ keV/MeV} \\ \text{QF}_{\alpha} = 0.1494 \pm 0.0002 \end{array} \right]
\]

\[
^{210}\text{Po} \left[ \begin{array}{c} \overline{LY}_{\alpha} = 3.011 \pm 0.003 \text{ keV/MeV} \\ \text{QF}_{\alpha} = 0.1813 \pm 0.0002 \end{array} \right]
\]

The increase with energy of the \(\overline{LY}_{\alpha}\) is responsible for the curvature of the \(\alpha\) band, an effect already observed by Refs. [11,12]. In Fig. 2 we show the result obtained fitting the \(\alpha\) band with a degree 2 polynomial.

We note that \(^{209}\)Bi and \(^{210}\)Po are internal contaminations of the crystal. In principle their LY could differ from that of a pure \(\alpha\) particle because a fraction of the total energy (about 2\%) is carried by the nuclear recoil (\(R\)). However, the difference in the LY of \(\alpha\) and \(\alpha + R\) events is far below our sensitivity. For this reason, here and in the following, we will assume \(\alpha\) and \(\alpha + R\) events as having the same LY. \(\alpha\) decays on the excited state of the daughter nucleus give rise to mixed \(\alpha + \gamma\) events. In this case, the light signal is higher than what is expected for a pure \(\alpha\) emission, producing therefore events that lie in between the \(\alpha\) and the \(\beta/\gamma\) band. An example is the \(^{210}\)Po decay, a contaminant responsible for the events appearing in the mixed \(\alpha + \gamma\) band, just at the left of the \(^{210}\)Po (pure \(\alpha\)) line. \(^{210}\)Po is produced in bismuth by thermal neutron interaction and it accumulates in the material due to its long half-life (see Ref. [11]). The isotope \(\alpha\) decays \((Q = 5036.4\) keV) to different excited levels of the daughter isotope \((^{206}\)Tl) with the contemporary emission of one or more \(\gamma\) rays. The two spots (visible in Fig. 2) are ascribed to the decays to the two lowest levels [with a sum branching ratio (BR) of 94.5\% and energies of 265.6 or 304.9 keV] and to two higher ones (with a BR of 1.4\% and 3.9\% and energies of 634.5 and 649.6 keV). Indeed, the probability of full containment of the emitted gammas is high, ranging from \((84.2 \pm 0.4)\%\) for the lowest energy \(\gamma\) to \((54.5 \pm 0.3)\%\) for the highest one. Taking into account the structure of the deexcitation cascades of the four levels, the intensities measured for the two spots agree with the tabulated branching ratios within 1 standard deviation [13].

\textbf{C.} \(^{209}\)Bi decay: Similar to \(^{210}\)Po is the case of our interest: \(^{209}\)Bi decay. It follows two different paths to the \(^{205}\)Tl ground state labeled I and II in Fig. 1. Process I produces an \(\alpha\) particle plus a recoil \((Q = 3137\) keV); being a monochromatic pure \(\alpha\) decay it should produce a line in the \(\alpha\) band (the probability of fully containing both the \(\alpha\) particle and the recoiling nucleus inside the crystal, \(\epsilon_{f}\), is obviously \(\sim 1\)). Process II produces an \(\alpha\) particle plus recoil \((Q = 2933\) keV and either a prompt ray \([E_{\gamma} = 204\) keV, containment efficiency \((92.1 \pm 0.5)\%\) or a

![Heat and Light Spectra](image-url)
Fig. 4 (color online). Zoom of the light versus heat scatter plot in correspondence to $^{209}$Bi events. The projections of the two spots corresponding to processes I and II along the heat axis (top panel) and the light axis (lateral panel) are shown. The horizontal axis reported on the top of the plot is obtained with a linear calibration based on the $^{209}$Bi 3137 keV pure $\alpha$ line. The circle indicates the position that should correspond to the $\alpha$ + recoil emitted in process II (2933 keV). Colors follow the convention used for Fig. 2.

conversion electron followed by its deexcitation x rays ($E_{\text{IC}} = 204$ keV, containment efficiency $\sim$100%) [13]. Taking into account the internal conversion coefficient of $^{205}$Tl ($\alpha_T = 0.46$, Ref. [14]), the probability that all the energy is deposited into the BGO crystal is $\epsilon_{\gamma} = (95.7 \pm 0.7)$%. We note also that the two processes of deexcitation of the 204 keV level (the gamma emission or the internal conversion) are virtually indistinguishable since (in the case of our interest, namely, that of full containment of the emitted particles) they correspond to the same energy deposition into the BGO crystal and in both cases the energy carriers are electrons or photons, and therefore characterized by the same LY. In such cases we have a monochromatic $\alpha + \gamma$ event that should produce a line in the mixed $\alpha + \gamma$ band.

This is exactly what we observe in our data: two spots corresponding to roughly the same heat position (3350 keV on the $\beta/\gamma$ calibration scale), but a different light emission, are clearly visible (see Fig. 2). Zooming the scatter plot in the $^{209}$Bi region (Fig. 4) we can observe that not only the light position of the two spots is different, but also the heat one. Indeed, although the emitted particles carry the same total energy, in process II a larger fraction of it is spent in the production of scintillation light. The difference in the light position is

\[
\text{Light}_{\text{II}} - \text{Light}_{\text{I}} = \gamma \cdot E_{\gamma} - \Delta \text{Light}_{\alpha},
\]

where $\Delta \text{Light}_{\alpha}$ is the difference between the light signal of the 3137 keV $\alpha + R$ of process I and that of the 2933 keV $\alpha + R$ of process II. This relationship can be used to estimate the energy of the emitted photon, further proving that we are observing $^{209}$Bi decay.

Indeed, since the nonlinearity of the heat axis is very small, we can linearly calibrate it in the proximity of the $^{209}$Bi peak (in Fig. 4 the heat axis calibrated in this way is drawn on the top). Using this calibration, we can obtain the heat position expected for an $\alpha + R$ event of 2933 keV. This results in (3133.8 $\pm$ 0.3) keV (the position is indicated with a circle in Fig. 4). Then, using the fit of the pure $\alpha$ band, we compute the value of $\Delta \text{Light}_{\alpha}$. Finally, we obtain for $E_\gamma$ a value of (192 $\pm$ 8) keV, validating our hypothesis.

The branching ratio for the two $^{209}$Bi decays are obtained fitting the light spectrum with the sum of two Gaussians having the same width and intensities: \((1 - \text{BR}) \cdot \epsilon_{\text{II}}\) for the intensity of the GS-ES transition and \(\text{BR} \cdot \epsilon_{\text{I}}\) for the intensity of the GS-GS transition. The result is a BR of (98.8 $\pm$ 0.3)% for the GS-GS transition. The total number of events corresponding to the decay is 2199 $\pm$ 66, with a detection efficiency of (87 $\pm$ 2)% (which accounts for both the trigger efficiency and the pulse-shape cuts efficiency). This yields a half-life for the $^{209}$Bi nucleus of $t_{1/2} = (2.01 \pm 0.08) \times 10^{19}$ yr. Finally the GS-GS transition partial width is $t_{1/2} = (2.04 \pm 0.08) \times 10^{19}$ yr, in good agreement with the previously reported one, Ref. [1].

IV. Conclusion.—This work provides a compelling evidence of the observation of $^{209}$Bi decay through the contemporary observation of the ground state and excited state transitions. While confirming the half-life of the isotope already measured by Ref. [1], we were also able to add new experimental information on $^{209}$Bi, namely, the branching ratio between the ground state and the excited state transition. This study covers a seldom explored field, namely, that of hindered $\alpha$ decays providing experimental inputs for a better development of the theoretical framework of nuclear models, Ref. [15]. In the mean time, these results prove once more the potentialities of the bolometric technique in the study of rare nuclear processes.

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The photon containment efficiencies quoted—or used—in this Letter are evaluated with a Monte Carlo simulation based on GEANT-4 (using the Livermore physics list). In the simulation photons are generated with a uniform distribution inside the bulk of a BGO crystal (a cube, 5 cm on the side). The containment efficiency is defined as the fraction of cases where the entire energy of the photon is deposited inside the crystal, the quoted error is statistical. In the case of $^{210}$Bi levels, the deexcitation of excited nuclear levels proceeds through a cascade that is reproduced in the simulation (with the contemporary generation of two photons) in order to account for the correct efficiency for each transition. For alpha particles, nuclear recoils, and the internal conversion process (yielding a conversion electron followed by an x-ray emission), we have assumed a containment efficiency of 1, as it is expected due to the short range of the emitted particles when compared with crystal size.


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This paper was published online on 9 February 2012 with an omission of an author byline footnote and an error in Fig. 4. L. Gironi’s byline footnote, “luca.gironi@mib.infn.it”, has been added, and the figure has been replaced online as of 22 March 2012. The figure is incorrect in the printed version of the journal; therefore, for the benefit of the print readership, the figure is duplicated below.

![Diagram]

**FIG. 4** (color online). Zoom of the light versus heat scatter plot in correspondence to $^{209}$Bi events. The projections of the two spots corresponding to processes I and II along the heat axis (top panel) and the light axis (lateral panel) are shown. The horizontal axis reported on the top of the plot is obtained with a linear calibration based on the $^{209}$Bi 3137 keV pure $\alpha$ line. The circle indicates the position that should correspond to the $\alpha +$ recoil emitted in process II (2933 keV). Colors follow the convention used for Fig. 2.