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Experimental detection of α-particles from the radioactive decay of natural bismuth

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The only naturally occurring isotope of bismuth, ^{209}Bi, is commonly regarded as the heaviest stable isotope. But like most other heavy nuclei abundant in nature and characterized by an exceptionally long lifetime, it is metastable with respect to α-decay[1]. However, the decay usually evades observation because the nuclear structure[2,3] of ^{209}Bi gives rise to an extremely low decay probability and, moreover, generates low-energy α-particles difficult to detect. Indeed, dedicated experiments[2–6] attempting to record the α-decay of ^{209}Bi in nuclear emulsions failed. However, scintillating bolometers[7–9] operated at temperatures below 100 mK offer improved detection efficiency and sensitivity, whereas a broad palette of targets could be available[10]. Here we report the successful use of this method for the unambiguous detection of ^{209}Bi α-decay in bismuth germanate detectors cooled to 20 mK. We measure an energy release of 3,137 ± 1 (statistical) ±2 (systematic) keV and a half-life of $(1.9 \pm 0.2) \times 10^{19}$ yr, which are in agreement with expected values.

Mass excess measurements show that ^{209}Bi, the only naturally abundant isotope of bismuth, should decay to the more stable thallium isotope ^{205}Tl (Fig. 1a,b). The low available decay energy Q_α induces a vanishing probability P_α for tunnelling of the α-particle through the nuclear potential barrier. Moreover, ^{209}Bi is next to doubly 'magic' nucleus ^{208}Pb ($Z = 82$; $N = 126$) and the need to pick nucleons across this extremely stable configuration, together with the poor overlap of initial and final nuclear states wave functions, further explain why the α-decay of ^{209}Bi is very rare, and thus difficult to detect.

Direct observation of the decay is also not easy: the α-particles move in solid matter over distances of around 10 μm (ref. 11), which are impractical to detect using common detectors (gas counters, semiconductors). It was thought that the appearance in 1945 of commercial nuclear emulsions that could be loaded with Bi would solve the technical issues of detection. Numerous experiments followed[2–6]: the exposure times were long, and those results found positive[5,6] were invalidated afterwards (Table 1).

We detected α-decay of ^{209}Bi using the scintillating bolometer technique. The earlier scintillating bolometers used CaF_2(Eu) and a silicon photodiode, glued on the crystal, as the light detector[9]. After recording an initial high-resolution α spectrum within a micro-target (a 1-mg diamond bolometer at 1.3 K, ref. 12), we introduced a 'heat-and-light' pair of bolometers, with 300 mg of scintillating CaF_2(Eu) at 130 mK (ref. 8). As part of our development of more massive scintillating bolometers (Fig. 1c) for the Dark Matter search experiment ROSEBUD, a 45.7-g BGO ($Bi_4Ge_3O_{12}$) bolometer exhibiting unusual discriminating properties at 20 mK was checked at Orsay for contamination at high energy (Fig. 2): this crystal had already shown a strong ^{207}Bi contamination[13]. Thanks to their high quenching factor, α-particles could be perfectly separated from events induced by cosmic rays (see Supplementary Information). An unexpected α line was observed at $E = 3{,}130 \pm 16$ keV, now attributed to ^{209}Bi decay. The publicly available SRIM-2003 package can be used to estimate the ranges in BGO of the decay products—respectively 7.9 μm for the α-particle ($E_\alpha = 3{,}077$ keV) and about 170 Å for the ^{205}Tl recoiling nucleus ($E_{Tl} = 60$ keV). These events are fully contained in our massive crystal, guaranteeing 100% detection efficiency. The associated half-life of ^{209}Bi is then calculated from the events rate to be $T_{1/2} = 1.44\text{–}1.95 \times 10^{19}$ years (68% confidence level, CL).

In all materials tested, bolometers register indiscriminately α-particles and their associated nuclear recoils with nearly equal

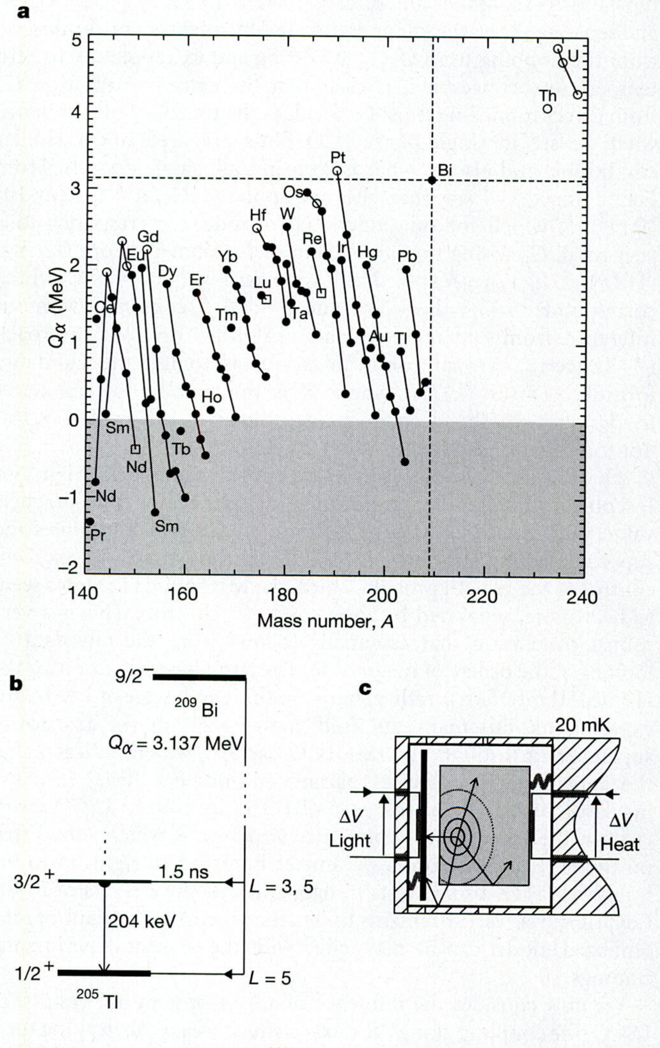

Figure 1 The bolometric detection of ^{209}Bi decay. **a**, Stability of heavy ($A > 140$) naturally abundant elements. The mass excess Q_α, calculated from tables[1], is given for elements found in average abundance >0.1 mg ton^{-1} in the Earth's crust. Lines connect isotopes of the same element. α-particle decay is energetically allowed only for $Q_\alpha > 0$ (out of the grey area). Squares, β-decay reported; black circles α-decay not yet observed; white circles, α-decay detected. Element (year of report): ^{144}Nd (1954); ^{147}Sm (1933); ^{148}Sm (1970); ^{152}Gd (1959); ^{174}Hf (1959); ^{186}Os (1975); ^{190}Pt (1921); ^{232}Th (1898); ^{234,235,238}U (1896). **b**, α-decay scheme of ^{209}Bi. Nuclear states are shown with their adopted spin and parity (J^π), energy level and half-life[27]. The finite quanta of angular momentum L, to be carried by the α-particle according to the transition rules, are indicated. **c**, Measuring heat and light in scintillating bolometers. Scintillating bolometer (BGO cylinder; diameter = height = 20.2 mm) and optical bolometer (germanium disk; diameter = 25 mm; 100 μm thick) face each other in an Ag-coated light-reflecting cavity. A continuous base temperature $T = 20$ mK—the key issue for high sensitivity—is provided by a ^{3}He–^{4}He dilution refrigerator, to which the detectors are thermally linked. Thermo-resistive sensors made from neutron transmuted doped germanium (NTD-Ge) are polarized at constant current and convert temperature rise into voltage signal. The voltage pulses (μV) that follow an interaction are amplified, digitalized and stored (see examples in Supplementary Information). Suspensions are not drawn. Small holes in the cavity allow external excitation, either with optical pulses sent through optical fibres, or with external radioactive sources.

Table 1 **Survey of attempts to detect ^{209}Bi decay**

Year of publication	Final level tested in ^{205}Tl	Detection technique	Effective exposure*		Number of reported (expected) events†
			Atoms × year	Time	
1949 (ref. 4)	Ground state	N.E.	4.3×10^{15}	≤7 months	<1 (0)
1951 (ref. 5)	Ground state	N.E.	7.0×10^{18}	2.25 years	18 (0)
1952 (ref. 6)	Ground state	N.E.	2.0×10^{18}	3.3 months	7 (0)
1958 (ref. 2)	Ground state	N.E.	2.9×10^{18}	3 years	<1 (0)
1972 (ref. 3)	Ground state	N.E.	2.3×10^{20}	"years…"	<1 (9)
2000 (ref. 20)	204 keV	$Bi_2O_3 + \gamma$ spectrometer	3.0×10^{21}	29.3 days	<46 (1–3)
2003 (this work)	Ground state	BGO bolometers	3.5×10^{21}	5 days	128
2003 (this work)	204 keV	46-g BGO bolometer	1.1×10^{20}	5 days	≤1 (0)

N.E.: Bi-loaded Nuclear Emulsion (Ilford type C2[2,4–6] or L4[3]): the energy of the α-particles is inferred from their range in photographic plates, with typical energy resolution 700 keV FWHM[26].

*Effective exposure (atoms × years) includes authors' own estimation of the detection yield, taking into account microscopist's efficiency, background level, absorption of gammas and so on. To obtain the published limit, or value, of $T_{1/2}$, multiply this number by 0.693 = ln(2) and divide by the number of reported events.

†The expected number of events is deduced from this work. The false-positive events reported in nuclear emulsions[5,6] might result from "chemical fog"[26], while, on the other hand, uncontrolled fading of plates could explain why detection was marginally overlooked in the longest exposure time experiment[3]. Updated data from the Oroville's experiment[20] were kindly provided by those authors.

thermal conversion yields[14,15] (see also discussion in Supplementary Information). Assuming the equality condition to be true also for BGO, we mounted and tested a second BGO bolometer (91.2 g), under external ^{241}Am irradiation (Fig. 3). The crystal was grown from purer raw material (Crismatec). Although its ^{207}Bi content was in fact strongly reduced (see spectrum in Supplementary Information), the former α line was still observed at a rate compatible with mass scaling, $T_{1/2} = 1.81\text{–}2.29 \times 10^{19}$ years (68% CL).

Combining the previous half-lives measurements increases statistical significance, to give $T_{1/2}(^{209}\mathrm{Bi}) = 1.9 \pm 0.2 \times 10^{19}$ years (68% CL), which we compared with the few published theoretical expectations[2,3]. We followed the later estimation[3] in which, as suggested by Rasmussen, the ^{209}Bi decay rate was calculated differentially from the decay rate of ^{210}Po ($T_{1/2} = 138.4$ days), on the basis of their nuclear structure similarities. After updating the relevant atomic masses and energy values, we revised the tabulated barrier penetration probabilities P_α (ref. 16,17), including the 'centrifugal potential' attributable to the finite angular momentum, and found an expected half-life $T_{1/2} = 4.6 \times 10^{19}$ years (formerly 1.1×10^{20} years) in close agreement with our experimental result.

The curvature of the α branch in Fig. 3, which showed increasing light emission with energy, implied at least some intrinsic non-linearity on the thermal channel as well, owing to conservation of energy. A slight opposite trend, which was indeed found on the heat channel, was easily corrected, as an accurate calibration through the well-identified α lines reduced the integral non-linearity below 10^{-3} over the whole spectrum (0–9 MeV). The energy of the new line, shifted by −12 keV in this process, was then calculated as $E = 3{,}137 \pm 1$ (stat.) ±2 (syst.) keV, precisely matching the

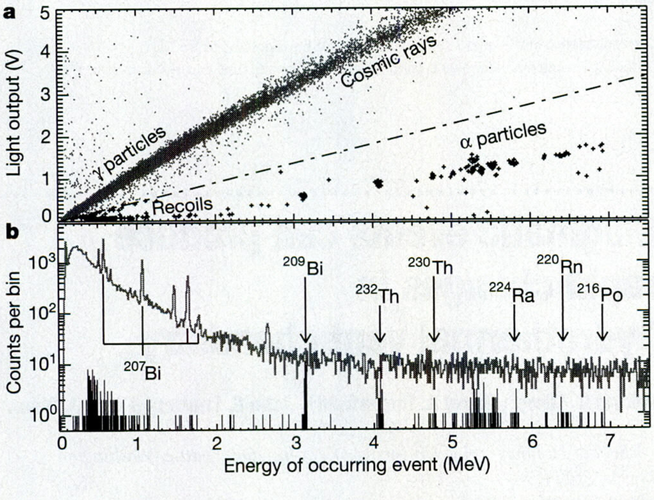

Figure 2 Discrimination of background events in a 46-g BGO bolometer. **a**, Background in the 46-g BGO bolometer (100 h). 'Light' signal versus 'Heat' signal. The latter has been converted into energy of occurring event in the BGO crystal. Two main classes of events are visible: a 'γ and cosmic rays' branch, giving high light signals owing to the high ionization power of these particles, and an α-particle branch. A third class of events is visible at the low-energy, low-light level: it is linked to nuclear recoils induced by ambient neutrons at laboratory level[13]. Separate calibration is needed[13,22]: ^{207}Bi lines from internal contamination are used for the γ branch; for the α branch, internal ^{232}Th, ^{230}Th, ^{224}Ra, ^{220}Rn and ^{216}Po α lines were identified step by step. Some were obviously paired events (see Supplementary Information); the final α calibration uses a weighted least-squares polynomial fit to these lines, including the 'zero energy' point. **b**, Energy spectra of alphas, recoils and gammas after application of a simple cut on light-to-heat signal ratio (dotted line in **a**). The expected location of all well-identified lines is reported, the γ spectrum being dominated by lines from ^{207}Bi ($T_{1/2} = 35.5$ yr; 3 Bq kg^{-1})[13].

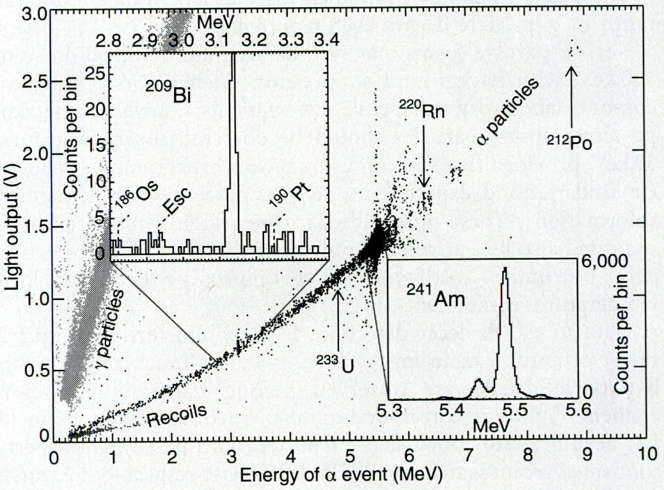

Figure 3 α events during ^{241}Am calibration in a 91-g BGO bolometer (114 h). α events (black dots) are selected according to their light-to-heat ratio (see Fig. 2). External ^{241}Am is used for calibration: the source is thin (activity ≈ 185 Bq cm^{-2}), and negligible energy loss in the source itself is assumed (see Supplementary Information). A low-rate, low-energy tail is visible, associated to the (back)scattering of α-particles in the support: these events allow for indisputable assessment of the newly detected line to α-decay, a unique feature of this technique. Events apart from the ^{241}Am lines were shown to be slight contaminations of the source itself: the associated events from ^{233}U, ^{220}Rn and ^{212}Po were used, together with the ^{241}Am and 'zero energy' lines, to provide, after a weighted least-squares polynomial fit, an accurate calibration of occurring α events (see details in Supplementary Information); their expected location is reported on the diagram. Right inset, spectrum of ^{241}Am. The fine structure is resolved, owing to the 18 keV FWHM resolution of the detector (the noise measured on the baseline has a negligible 6-keV contribution to this width). The same spectrum looked at from the light channel would show a 295-keV FWHM resolution only. Left inset, the ^{209}Bi α-decay line. Spectrum of recorded events (2.8 MeV to 3.4 MeV) during the 114-h ^{241}Am calibration run. The ^{209}Bi line stands out well from the background (around seven events estimated under the line). The location of conceivable α-decays in this energy window is indicated: ^{186}Os, ^{190}Pt, and ^{209}Bi decay to ^{205}Tl 204 keV level γ escape peak (Esc).

expected value of ^{209}Bi α-decay, $Q_\alpha = 3{,}137 \pm 0.9\,\mathrm{keV}$ (ref. 1, see comment in Supplementary Information).

Finally, possible contaminations by already-known α emitters of close Q_α were considered: ^{186}Os ($T_{1/2} = 2.0 \times 10^{15}$ years; $Q_\alpha = 2{,}822\,\mathrm{keV}$) and ^{190}Pt ($T_{1/2} = 6.5 \times 10^{11}$ years; $Q_\alpha = 3{,}249\,\mathrm{keV}$) were discarded because the results implied very high atomic concentrations, respectively Os/Bi ≈ 6,500 p.p.m. and Pt/Bi ≈ 250 p.p.m. A dedicated quantitative Pt implantation experiment in BGO, followed by a secondary ion mass spectrometry (SIMS) analysis, proved that Pt concentration was under 2 p.p.m. Higher concentrations were very unlikely[18], and would have strongly affected the scintillating properties of the crystal[19].

The auxiliary observation of ^{209}Bi decay to the first excited state of ^{205}Tl (Fig. 1b) would constitute a conclusive test of our proper identification of the α line. Such a—negative—search was performed recently, using a very low background facility under the Oroville dam, where 204 keV γ-particles emerging from 1.8 kg of Bi_2O_3 were looked for[20]. Thanks to the high Z of Bi, and to their slow time constant ($\tau \approx$ a few ms $\gg$ 1.5 ns), BGO bolometers naturally sum the energies of the α-particle and of the following absorbed γ, adding another event to the 'full energy' peak. However, the γ escape probability is not negligible: we estimate it to be 12.8% in our 46-g BGO detector by Monte Carlo simulations. Taking into account internal conversion[21], the overall efficiency for detecting α-decay at this level through recording of the escape peak is $\eta = 8.8\%$. With reasonable assumptions (see Supplementary Information), the associated partial half-life is estimated to be $T_{1/2} = 6\text{–}13 \times 10^{20}$ years: a two-month exposure would then probably lead to its detection (Table 1).

These data extend by more than three orders in magnitude the range of α-particle decays ever reported ($T_{1/2} = 7 \times 10^{15}$ years; ^{148}Sm). α-particle events may now be accurately tracked down to 500 keV, with background suppression efficiency over 98%, in common laboratory radioactive environments. Equivalent rejection power against γ-particles should be observed down to around 50 keV, provided that the confusing recoils background is reduced (in underground experiments, for example; see Supplementary Information). These possibilities in the search for rare events—indicated also by our recent improvements of the ^{180}W α-particle decay rate limit[22]—justify the greater complexity added by the low-temperature requirements.

With this ^{209}Bi decay detection, BGO bolometers open up the rarest of natural main energy lines—whereas much rarer double β-particle decays are observed among naturally abundant elements[23], no line corresponding to the neutrinoless mode $0\nu\beta\beta$ has been hitherto convincingly reported. This particular property motivated recent searches for ^{209}Bi decay: with respect to the search for proton decay, a proposal was made to use 'freshly produced' bismuth to check for deviation of radioactive decay from the exponential law at small times[24,25]. ☐

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Magmatic events can produce rapid changes in hydrothermal vent chemistry

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The Endeavour segment of the Juan de Fuca ridge is host to one of the most vigorous hydrothermal areas found on the global mid-ocean-ridge system, with five separate vent fields located within 15 km along the top of the ridge segment[1]. Over the past decade, the largest of these vent fields[2], the 'Main Endeavour Field', has exhibited a constant spatial gradient in temperature and chloride concentration in its vent fluids, apparently driven by differences in the nature and extent of subsurface phase separation[3]. This stable situation was disturbed on 8 June 1999 by an earthquake