ISOTTA ISOTope Trace Analysis

Advanced Techniques for the Production, Purification and Radio-Purity Analysis of Isotopically Enriched Sources for Double Beta Decay

Project coordinator

CNRS

Andrea Giuliani - Centre National de la Recherche Scientifique, CSNSM, Orsay, France

Co-applicants

CNRS (France)

INFN (Italy)

NCBiR (Poland)

Mathieu Bongrand – Centre National de la Recherche Scientifique, LAL, Orsay, France Oliviero Cremonesi – Istituto Nazionale di Fisica Nucleare, Sez. di Milano-Bicocca, Italia Ioan Dafinei – Istituto Nazionale di Fisica Nucleare, Sez. di Roma1, Italia Fernando Ferroni – Università la Sapienza, Roma, Italia Xavier Garrido - Université Paris-Sud 11, LAL, Orsay, France Jan Kisiel – University of Silesia, Katowice, Poland

Jerzy Wojciech Mietelski - H. Niewodniczanski Institute of Nuclear Physics, Krakow, Poland

Fabrice Piquemal - Centre National de la Recherche Scientifique, CENBG, Bordeaux, France

Stefano Pirro – Istituto Nazionale di Fisica Nucleare, Sezione di Milano-Bicocca, Italia

Ezio Previtali - Istituto Nazionale di Fisica Nucleare, Sezione di Milano-Bicocca, Italia

Xavier Sarazin - Centre National de la Recherche Scientifique, LAL, Orsay, France

Laurent Simard – Université Paris-Sud 11, LAL, Orsay, France

Jacek Szabelski – Andrzej Sołtan Institute for Nuclear Studies, Łodz, Poland

Marcin Wojcik - Institute of Physics, Jagiellonian University, Krakow, Poland

Agnieszka Zalewska – H. Niewodniczanski Institute of Nuclear Physics, Krakow, Poland

Associated partners

Ukraine

Russia

Germany

Kai Zuber – Technische Universität Dresden, Germany
Fedor Danevich – Institute for Nuclear Research, Kyiv, Ukraine
Roman. S. Boiko – Institute for Nuclear Research, Kyiv, Ukraine
Ruslan Podviyanuk – Institute for Nuclear Research, Kyiv, Ukraine
Vladimir Tretyak – Institute for Nuclear Research, Kyiv, Ukraine
Victor Brudanin – Joint Institute for Nuclear Research, Dubna, Russia
Dmitry Filosofov – Joint Institute for Nuclear Research, Dubna, Russia
Oleg I.Kochetov – Joint Institute for Nuclear Research, Dubna, Russia
Evgeny Yakushev – Joint Institute for Nuclear Research, Dubna, Russia

ISOTTA in a nutshell



Guidelines for the procurement of 1 ton of radiopure isotope for 0v-DBD within 3 years from the start

ISOTTA aims at:

- reviewing the existing isotope producers
- investigating new enrichment technologies
- identifying purification procedures
- developing techniques able to test the radiopurity of enriched samples at the level of few μBq/kg prefiguring final detectors



coordinated approach to the isotope problem, joining synergically the expertizes of the main European actors in the field

Current challenge in neutrinoless Double Beta Decay search

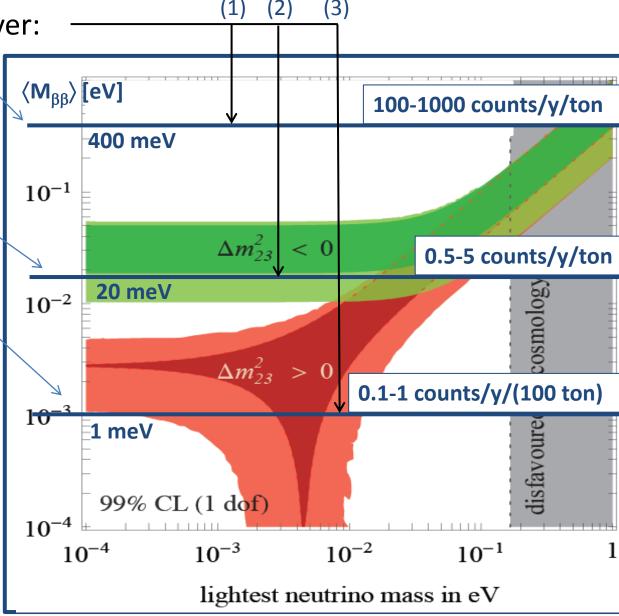
Three hurdles to leap over:

First: scrutinize **Klapdor** and **approach inverted hierarchy region** (EXO-200, GERDA, CUORE, SuperNEMO, NEXT, KamLAND-ZEN, SNO+...)

Second: cover inverted hierarchy region (technically feasible but today no planned experiment can credibly do it)

Third: attack direct hierarchy region (beyond the reach of every conceivable technique at the moment)

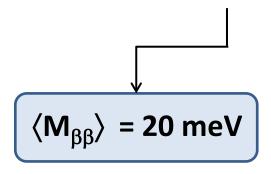
ISOTTA aims at giving a substantial contribution to the second step



Necessity of enrichment at 1-ton scale

Evaluation of the **expected signal rate** for several interesting isotopes (**counts /(5 y ton**) for three different nuclear calculations, **assuming**:

Candidate	pnQRPA	IBM-2	ISM
⁷⁶ Ge	4.0	7.9	1.7
¹³⁶ Xe	4.8	12	3.9
¹³⁰ Te	9.4	17	5.6
¹¹⁶ Cd	12	/	/
⁸² Se	7.3	21	6.9
¹⁰⁰ Mo	10	19	/
¹⁵⁰ Nd	/	24	/



(approximately, lower bound of the inverted hierarchy region)

Just to have enough signal

ultrapure isotopically enriched material at the level of 1 ton is necessary

ISOTTA objectives

Main objective

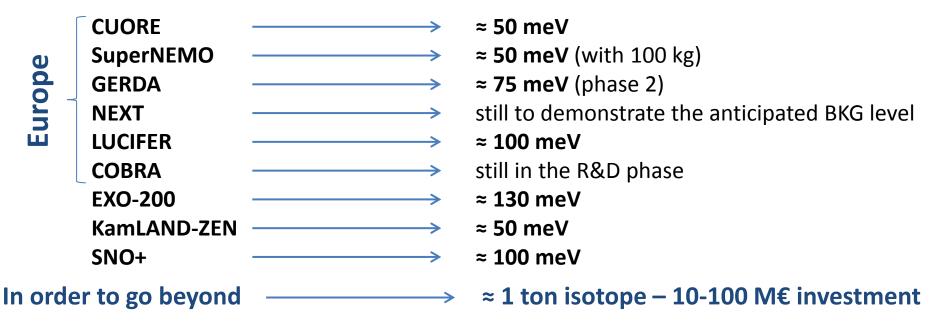
radio-purity of the final source) of a large amount (at the 100 kg – 1 ton scale) of isotopically enriched material for the performance of a next generation 0v-DBD experiment.

achieved through the following intermediate goals:

- Overview of the isotope producers
- Overview of possible innovative technologies for the isotope production
- Procurement of samples of isotopically enriched materials
- Development of techniques and facilities for the isotope radiopurity characterization with
 - **standard methods** (nuclear and mass spectroscopy)
 - **innovative methods**, prefiguring the structure of future 0v-DBD experiments: enriched materials already cast in the form of the sources foreseen for the future detectors (bolometric absorbers, thin foils, scintillators and semiconductors)
- development of purification methods by using combination of chemical (recrystallization) and physical (vacuum distillation, filtration, zone melting) approaches

Urgency of the research proposed in ISOTTA

Current experiments, running or in construction, do not have the sensitivity to really attack the **inverted hierarchy region**:



It cannot be done without previous careful investigation of the enrichment process /purification methods / radiopurity level

ISOTTA proposes to start the preliminary work now in order to be ready within three years (when current experiments will have shed light on future approaches)

ISOTTA strategy (1)

1 Focus the attention on **several** very interesting candidates

Q-value higher than 2615 keV (out of the bulk of gamma radioactivity)

Can be studied with:

- **Bolometric technique** (high energy resolution ≈ 3-5 keV FWHM)
- External source (tracko-calo) approach (full event reconstruction)

Only relevant $\beta^+\beta^+$ candidate

- 2 Procure samples of isotopes in the ISOTTA consortium
 - European Bank of Isotopes established inside ILIAS-FP6
 - Samples belonging to
 - the associated partner **Dubna**
 - the associated partner **Kiev**
 - the **CUORE** collaboration
 - the **LUCIFER** collaboration

ISOTTA strategy (2)

3 Acquire small samples of isotopes with ISOTTA funding

- Develop and coordinate trace analysis techniques
- pre-screening investigation (ICP-MS, gamma/beta spectrometers)

 final investigation
 BiPo detectors
 Calorimetric detectors (scintillators charge-collection devices)
 (Scintillating) bolometers

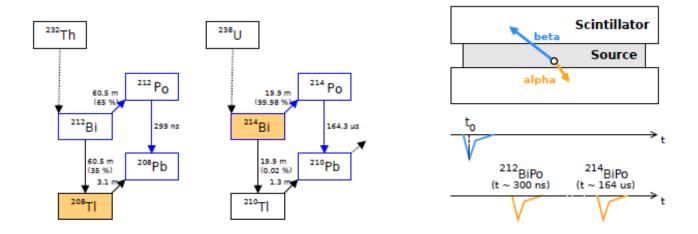
Sensitivity at the level of a **few** μ **Bq/kg** for the isotopes under study

⑤ Develop and coordinate purification techniques

Sensitivities of the advanced techniques proposed in ISOTTA

External source approach – the BiPo way

Principle: ²¹⁴Bi and ²⁰⁸Tl contaminations in sources in the form of thin sheets measured by **BiPo processes** from natural radioactivity chains ———— **beta + delayed alpha**



Sensitivity: from preliminary measurements, conservative estimation of BiPo3 detection limits

Example: **1.3 kg of ⁸²Se** thin source

$$A(^{208}TI) < 6.2 \mu Bq/kg 90 %CL$$

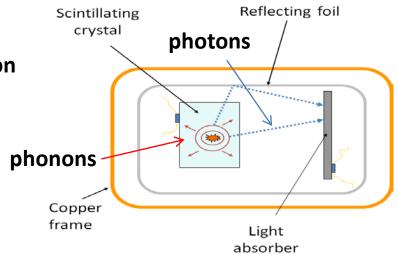
 $A(^{214}Bi) < 20.2 \mu Bq/kg 90 %CL$

Sensitivities of the advanced techniques proposed in ISOTTA

2 "Self counting" approach – the scintillating bolometer way

Principle: a crystal containing the isotope under study is operated as scintillating bolometer

Simultaneous detection of **heat** and **scintillation** light and different quenching factor allow excellent **alpha / beta separation**



Sensitivity: example – **100** g of ¹⁰⁰Mo operated inside a scintillating bolometer

- 0 background above 2615 keV in the alpha region in one month

 $A(^{232}Th)$, $A(^{238}U)$ < 11 μ Bq/kg 90 %CL

Since the main alpha lines of the natural radioactive chains are individually detected with high energy resolution, breaking of secular equilibrium can be directly observed

Sensitivities of the advanced techniques proposed in ISOTTA

The techniques adopted in *ISOTTA* can provide unprecedented sensitivities at the level of

a few μBq/kg

for several DBD candidates and for the most dangerous isotopes

- ²³⁸U, ²³²Th and their daughters
- Specifically, ²⁰⁸TI and ²¹⁴Bi
- Cosmogenic nuclide contribution (directly appreciable with the "self counting" approach)

level required by DBD experiment aiming at attacking the **inverted hierarchy region**

The structure of ISOTTA

5 work-packages

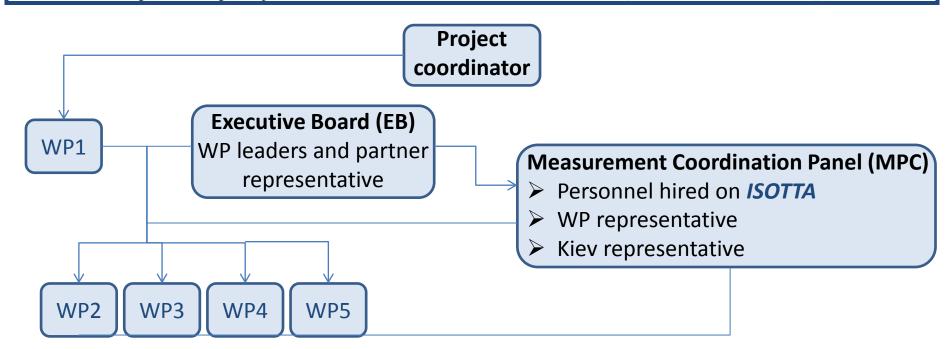
WP1: Management and Coordination [Andrea Giuliani, CNRS]

WP2: Isotope production and purification [Ezio Previtali, INFN]

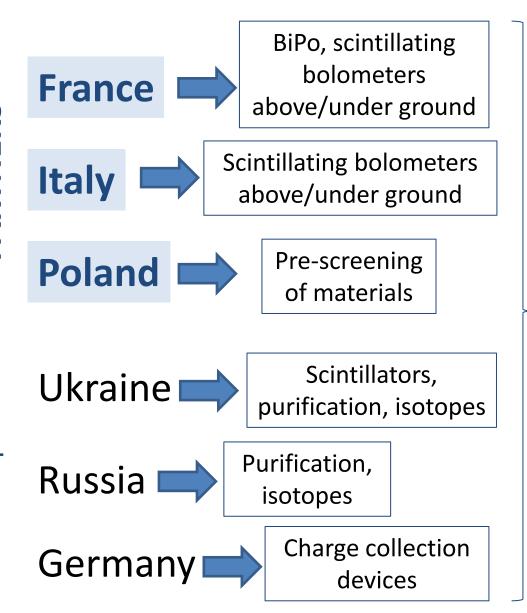
WP3: Isotope radio-purity assessment with nuclear and mass spectroscopy [Marcin Wojcik, IF UJ]

WP4: Isotope radio-purity assessment with external source approach [Laurent Simard, CNRS]

WP5: Isotope radio-purity assessment with calorimetric methods [Stefano Pirro, INFN]



Added value of transnational collaboration



Distributed laboratory with a few µBq/kg sensitivity

WP1: Management and Coordination

Objectives:

- monitor the technical and scientific work of the other Work Packages;
- guarantee that the proposed schedule is maintained, that the project milestones are achieved and that the deliverables are produced on time;
- organize and coordinate the general meetings;
- coordinate the activities proposed by the MCP;
- prepare the three general ISOTTA annual reports (which will emphasise the inter-comparison results and the activities which cut the boundaries between WPs);
- organize the dissemination program in collaboration with the EB;
- coordinate the protection, use and dissemination of the knowledge generated during the period of the
 project, establishing a light website that will work as a repository for the documentation (reports,
 presentations, databases, protocols) produced by the consortium;
- provide the contact and the communication with the ASPERA Common Call Secretariat.

Deliverables:

- Three annual reports
- Organization of the general meetings of ISOTTA and related material

Budget

Institution	Requested	Assigned
CNRS	164000	164000
INFN	130000	54000
NCBiR	268700	~160000 (ask A.Z.)

Milestones and deliverables (WP1, WP2)

	Description of Deliverable / Milestone	Month
M1.1	Set up of the Measurement Coordinating Panel	2
D1.1	Organization of the general meeting and related documentation	4)10,16,22,28,34
D1.2	Website	6
D1.3	Annual reports (within inter-comparison among isotopes and technologies)	12)24,36
M2.1	Overview of the isotopes available inside the consortium	2
M2.2	Identification of the isotope producers to be contacted	4
D2.1	Report with overview of the producers and identification of the plants	6
D2.2	Preparation of a sequence of small enriched samples	10,22
D2.3	Comparison between different enrichment techniques	12,24
D2.4	Report on the chemical and physical characteristics of the prepared samples	30
M2.3	Selection of the most reliable isotope producers	32
D2.5	Identification of purification procedures for the enriched materials	34
D2.6	Preparation of a standard production protocol for isotope enrichments	36

Table 2 - Time plan of ISOTTA

Month	2	4	б	8	10	12	14	16	18	20	22	24	26	28	30	32	34	36
WP1	M1	D1	D2		D1	D3		D1			D1	D3		D1			D1	D3
WP2	M1	M2	D1		D2	D3					D2	D3			D4	М3	D5	D6
WP3					D1	D2 D3 D4	M1				D1	D4 D5			M2		D1	D6
WP4	M1				D1		D2		D3	M2							D4	
WP5		M1			D1	D2			D3			D1		D2		D3	M2	

Milestones and deliverables (WP3, WP4,WP5)

	Description of Deliverable / Milestone	Month
D3.1	Report on the screening campaign of the enriched samples and sources	10)22,34
D3.2	Report on the MC results on expected performance of different a spectrometers	12
M3.1	Selection of the technology for a new large surface $\boldsymbol{\alpha}$ spectrometer	14
D3.3	Design study of a new Ge spectrometer with different shielding options	12
D3.4	Report on simulations of background for the α and γ spectrometers at SUNLAB	12,24
D3.5	Working prototypes of α and γ spectrometer and related report	24
M3.2	$lpha$ and γ spectrometers ready for measurements	30
D3.6	Report on performance of $\ \alpha$ and γ spectrometer and related measurements	36
M4.1	BiPo-3 detector available	2
D4.1	Report on the background of the BiPo-3 detector	10
D4.2	Technical report on the performance of the BiPo-3 detector	14
D4.3	Report on the internal radioactivity of the 82Se source	18
M4.2	Validation of the 82Se source	20
D4.4	Report on all the measurements made with the BiPo-1 and BiPo-3	34
M5.1	Aboveground set-ups ready for characterization of calorimetric detectors	(4
D5.1	Technical report on the performance of the calorimetric detectors	10,24
D5.2	Report on the internal radioactivity of natural crystalline samples	12, 28
D5.3	Report on the internal radioactivity of enriched crystalline samples	18,32
M5.2	Ranking of isotopes for a future large calorimetric 0v-DBD experiment	34

Month	2	4	6	8	10	12	14	16	18	20	22	24	26	28	30	32	34	36
WP1	M1	D1	D2		D1	D3		D1			D1	D3		D1			D1	D3
WP2	M1	M2	D1		D2	D3					D2	D3			D4	М3	D5	D6
WP3					D1	D2 D3 D4	M1				D1	D 4 D 5			M2		D1	D6
WP4	M1				D1		D2		D3	M2							D4	
WP5		M1			D1	D2			D3			D1		D2		D3	M2	