Review

Dark Matter Searches Using NaI(Tl) at the Canfranc Underground Laboratory: Past, Present and Future

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Abstract: Sodium Iodide Thallium doped (NaI(Tl)) scintillation detectors have been applied to the direct searches for dark matter since the 1980s and have produced one of the most challenging results in the field—the observation by the DAMA/LIBRA collaboration of an annual modulation in the detection rate for more than twenty cycles. This result is very difficult to reconcile with negative results derived from other experiments using a large variety of target materials and detection techniques. However, it has been neither confirmed nor refuted in a model independent way up to the present. Such a model independent test of the DAMA/LIBRA result is the goal of the ANAIS-112 experiment, presently in the data taking phase at the Canfranc Underground Laboratory in Spain. ANAIS-112 design and operation leans on the expertise acquired at the University of Zaragoza in direct searches for Dark Matter particles using different targets and techniques and in particular using NaI(Tl) scintillation detectors for about thirty years, which are reviewed in the first section of this manuscript. In addition to presenting the status and more recent results of the ANAIS-112 experiment, open research lines, continuing this effort, will be presented.

Keywords: dark matter; NaI(Tl) scintillators; annual modulation; DAMA/LIBRA puzzle; Laboratorio Subterráneo de Canfranc

1. Introduction

For almost a century, evidence coming from the Universe at different scales points to the existence of an unknown matter component in galaxies, clusters of galaxies and cosmological scales. The ΛCDM cosmological model satisfactorily explains the observations at the cost of introducing 84% of the matter in the form of Dark Matter (DM) and 68% of the energy in the form of Dark Energy (DE) [1,2]. Proposed alternative theories based on modified gravity or dynamics are not capable, at this moment, of reproducing observations without DM or DE.

Understanding the nature of the DM has been shown to be a challenge requiring complementary approaches from the different points of view of cosmology, astronomy,

astrophysics, and nuclear and particle physics (theoretical and experimental). Despite the large efforts invested in the last decades following different strategies (searches at accelerators [3], indirect [4] and direct searches [5–7]), no clues on the nature of DM are yet at hand.

In the context of direct DM searches, the goal is to measure the small energy deposited in the target material by the interaction of the DM particles filling the Milky Ways's halo at the Solar System position. The expected signal is strongly dependent on the particle DM model (mass, kind and magnitude of the coupling to quarks and leptons), and on the galactic halo model (density of DM and velocity distribution function, both at the Solar System position). Those models' parameters are either unknown or affected by large uncertainties. In addition, those parameters interfere with the target nuclei properties in such a way that comparison between results from experiments using different absorber materials are model-dependent. The low interaction rates of these DM particles imply that backgrounds have to be minimized by operating the experiment underground, by using radiopure materials in the detector building, by shielding conveniently (passively and/or actively) against external backgrounds, and profiting from any background rejection procedure at hand. After more than three decades of DM direct searches, most of the experimental results are compatible with the estimated backgrounds. At present, there are two relevant results requiring further understanding: the XENON-1T experiment observes an excess of events that can be attributed either to tritium contamination or exotic particles interactions (axions, neutrino magnetic moment, warm dark matter, etc.) [8,9]; and DAMA/LIBRA result in annual modulation that lasts for more than two decades and cannot be explained by any known background (see below).

For a positive identification of the DM in direct detection searches it is necessary to analyze distinctive features of a DM signal against backgrounds. This can be done by studying the annual modulation effect in the DM interaction rates [10] produced by the orbital motion of the Earth around the Sun along the year. The corresponding variation in the relative velocity of the DM particles with respect to the target nuclei induces a modulation with a one-year period and a maximum at about the 2nd of June in the DM interaction rate, for the standard halo model.

Sodium Iodide Thallium doped (NaI(Tl)) scintillation detectors are a convenient choice for detecting radiation in many applications. In particular, they have been applied to the direct searches for dark matter since the 1980s and have produced one of the most challenging results in the field—the observation by DAMA/LIBRA collaboration of an annual modulation in the detection rate for more than twenty years [11–16]. This result is very difficult to reconcile with the negative results derived from other experiments using a large variety of target materials and detection techniques [17–33]. However, it has neither been confirmed nor refuted in a model independent way up to the present. The DAMA/LIBRA signal is fully compatible with the annual modulation associated to DM particles distributed in our galaxy in a halo with standard properties, and all the systematic effects proposed to contribute to the signal have been discarded by the DAMA/LIBRA collaboration. It is important to solve this experimental puzzle, which could be the door into new physics, and in order to do so a model independent test needs to be faced. Although the DAMA/LIBRA result is incompatible with the rest of the experiments searching for DM in most of the analyzed scenarios, it should be stressed that this comparison is model dependent and the DM nature is still fully unknown.

A model-independent test of DAMA/LIBRA result requires an experiment using the same target material, a similar background and threshold energy to those of DAMA/LIBRA, large exposure (product of mass and measurement time) and stability, in order to analyze the annual modulation effect. This is the goal of the ANAIS-112 experiment, presently in the data taking phase at the Canfranc Underground Laboratory, in Spain. The ANAIS-112 design and operation leans on the expertise acquired at the University of Zaragoza using NaI(Tl) scintillation detectors for about thirty years, which is reviewed in this manuscript and has been collected in several PhD dissertations [34–41].

Other experiments all over the world share the ANAIS-112 goal. The COSINE-100 experiment is also in the data taking phase at the YangYang laboratory [42,43], in the Republic of Korea, with an effective mass of about 60 kg, instead of the 112.5 kg of ANAIS-112, and the 250 kg of DAMA/LIBRA detector. SABRE [44], PICOLON [45] and COSINE-200 are still in R&D phase, aiming at improving the radiopurity of the NaI(Tl) crystals, but using similar technology. COSINUS is also in the R&D phase, but using a completely different detection approach, based on the simultaneous measurement of the light and phonons produced following a particle interaction in the NaI(Tl) crystal, which allows identifying the particle by the different sharing between the two energy conversion channels [46]. Previously, DM-Ice17, operating at the South Pole, tried to address this goal, but its sensitivity was too low [47].

Thallium doped sodium iodide is a high-performing scintillating material: large size crystals can be grown (up to tens of kilograms) and the amount of light produced by unit of energy deposited is one of the highest among scintillating materials, at the level of 40,000 photons/MeV. The wavelength of the light produced matches perfectly with the peak sensitivity of bi-alkali PMTs, easing the fabrication of scintillation detectors. In the application to DM searches, the 100% non-zero spin of both component nuclei, ^{127}I and ^{23}Na, makes NaI a very interesting target for the case of spin-dependent interacting DM particle candidates, and the combination of heavy and light nuclei allows a good kinematical matching in a wide range of DM particle masses.

However, the radioactive background level achieved by NaI(Tl) detectors has practically not improved in the last twenty years. There are several reasons behind, on the one hand, raw powder purification and crystal growing technology of BICRON/Saint Gobain at the end of the last century was not replicated by other companies, and further efforts to go beyond that level date from the last decade. On the other hand, the use of PMTs for the light readout of NaI scintillation light allowed the rise of the technology, but it is at present a drawback to increase its sensitivity. PMTs are not radiopure enough and contribute strongly to the radioactive background budget of the detectors. Moreover, they produce spurious events interfering with the bulk scintillation light detection in a way which has shown to be very tough to keep under control, limiting the detection of small amounts of light and, as a consequence, further reduction of the achievable energy threshold. Because of that, other light detectors coupled to the NaI scintillating crystals are under study in order to improve the sensitivity of this detection technology.

The layout of the manuscript is as follows: Section 2 summarizes the Canfranc Underground Laboratory history; Section 3 focuses on those experiments using NaI(Tl) as target material applied to the search for DM; Section 4 reviews the status and prospects of ANAIS-112 experiment, aiming at testing the DAMA/LIBRA result on annual modulation with more than three-sigma confidence level; and Section 5 sets the scene for the continuation of these efforts in the next years and the prospects on the mid- and long-term.

2. The Old Canfranc Underground Facilities and the New LSC

The Canfranc Underground Laboratory is a singular facility providing an ultra-low radioactivity environment and therefore, enabling the realization of experiments searching for very rare processes. Since the end of the 1980s, the underground facilities at the Canfranc tunnel have played a relevant role in the field of Underground Physics, hosting leading experiments in neutrinoless double beta decay ($0\nu\beta\beta$) searches [48–52], particle dark matter searches [53–72] and solar axion searches using crystalline targets [59,73]. Many other results were derived from these efforts as by-products [74–94]. For more than twenty years, the University of Zaragoza was responsible of the operation and management of the Canfranc tunnel facilities, thanks to the work of the researchers of the group in Nuclear and Astroparticle Physics, now integrated in the Center for Astroparticle and High Energy Physics (CAPA).

In 1985, researchers of the University of Zaragoza, led by Prof. Ángel Morales and Rafael Núñez-Lagos, were determined to find a convenient underground location which

could open the door to operating their detectors in ultra-low radioactive background conditions. They followed an international trend towards this new field of research: new experiments were proposed and designed to increase the sensitivity for particle DM and $0\nu\beta\beta$ searches, which succeeded in a few years with improvements in the sensitivity of several orders of magnitude.

The two best locations found for an underground laboratory were a salt-mine in Remolinos (near Zaragoza) and the Somport tunnel, in the railway connecting Spain and France under the Pyrenees, in Canfranc, which has not been in use since 1970. The latter location was finally chosen because of its better environmental conditions: a lower amount of dust in the air and better natural ventilation. Lab 1 (see Figure 1) was readily conditioned for immediate operation, profiting from the existence of two small halls on both sides of the tunnel (10 m² each), at 780 m from the Spanish tunnel entrance. A few years later, Lab 2 was commissioned: a prefabricated cabin (about 15 m²) that could be moved onto the rails. It was first placed besides Lab 1, but afterwards, around 1991, it was joint to a second prefabricated module (27 m²) and moved towards higher rock overburden, at 1200 m from the tunnel entrance (see Figure 1).
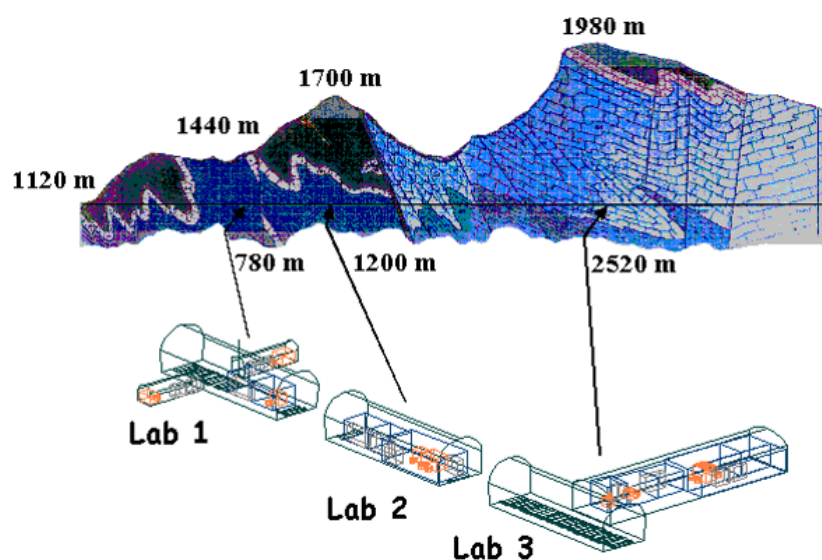

Figure 1. Scheme of the different experimental locations at the old Canfranc Underground Laboratory, in use up to 2010, except Lab 3, which is still in operation (now known as Lab2500). Distances are measured from the Spanish entrance to the Somport tunnel and rock overburden for each location is also shown.

In 1994, the excavation of a new road tunnel in the frame of a large infrastructure effort (highway from Somport to Sagunto) offered the opportunity of enlarging the underground facilities. A new experimental hall of 118 m² was excavated and conditioned for operation (Lab 3), profiting from the maximum rock overburden the mountain profile allows (see Figure 1).

Figure 1 shows a scheme of the different experimental halls available at the Canfranc Underground Laboratory before 2010. Distances are measured from the Spanish entrance to the Somport tunnel (left side in the figure), and the rock overburden corresponding to each location is shown. Corresponding values, in meters of water equivalent (m.w.e.), are for Labs 1, 2 and 3, respectively, of 675, 1380 and 2450 m.w.e.

Starting in 2004, a new excavation to build evacuation galleries connecting the railroad tunnel with the highway tunnel allowed us to project a new enlargement of the underground facilities. This new laboratory, the only underground facility in Spain, was declared a Spanish Unique Scientific and Technical Infrastructure (ICTS) and was established as a Consortium of the Spanish Ministry of Science and Innovation, the Aragon Regional

Government and the University of Zaragoza. In March 2006, the new facilities were inaugurated. In March 2007, the underground facilities closed because of important structural problems in the roofs of the experimental halls. Reparation works lasted until 2010, when the new facilities re-opened and access to researchers was granted again. The new Canfranc Underground Laboratory (see Figure 2) consists of more than 1000 m^2 of underground space, distributed in several experimental halls, and includes a clean room class 10,000 and mechanical workshops to support the experiments [95]. Additionally, ultra-low background services (such as HPGe detectors for gamma spectroscopy or a radon abatement system) and a copper electroforming facility are offered to the users [96]. An external scientific committee is in charge of reviewing the progress of the different experiments and the evaluation of the expressions of interest. The present scientific program of LSC is focused on the study of neutrino properties (hosting experiments devoted to the investigation of the double beta decay using different techniques, CROSS [97] and NEXT [98], and collaborating in the SuperKamiokande-Gd and HyperKamiokande projects) and on the direct detection of dark matter (with the TREX-DM experiment [99] and the DArTinArDM detector [100], together with ANAIS-112). Additionally, experiments and projects related to Geophyiscs, Biology, and studies on underground muon and neutron fluxes are underway.
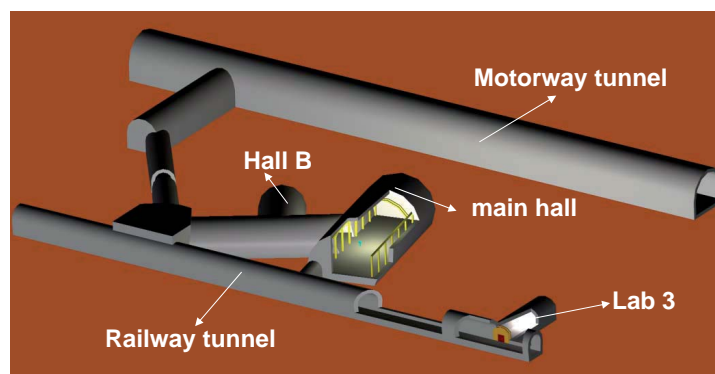

Figure 2. Scheme of the different experimental halls at the current Canfranc Underground Laboratory. Lab 3 is now called Lab2500. Main laboratory facilities (Lab2400) includes Hall A (main hall), Hall B, clean room, mechanical workshop and other services for the experiments.

3. Direct DM Searches at the Canfranc Underground Laboratory Using NaI(Tl)

3.1. Experiment DM32

At the end of the 1980s, the group of Nuclear and Astroparticle Physics of the University of Zaragoza owned 14 large sodium iodide (thallium doped) detectors, 10.7 kg each, at the Canfranc Underground Laboratory. They were originally intended for the selection of events coincident with a central detector, in a honeycomb configuration, in a search for ^{76}Ge $\beta\beta$ decay to excited state 2^+ at the Fréjus tunnel [101,102]. They were later used at the Canfranc Underground Laboratory, first, to discard that the Fréjus result [102] was a neutrinoless double beta decay detection [48], and after, to improve in several orders of magnitude the current limits at the time on the lifetime of $\beta^+\beta^+$ and β^+-EC processes, by searching the decay of ^{78}Kr [49,103]. The latter experiment was carried out in collaboration with the Institute of Nuclear Research (INR), Russia. At the beginning of the 1990s, a new experiment searching for DM using some of them as a target was proposed: DM32.

DM32 was a pioneer study of the annual modulation in the DM signal. It consisted of three modules 10.7 kg each of NaI(Tl), amounting to 32.1 kg of mass, and it was proposed as a proof of concept to estimate the achievable sensitivity in DM searches. The three modules were installed in a shielding consisting of 6 mm of Oxygen-Free High thermal Conductivity (OFHC) copper, 20 cm of low-activity lead, a PVC box kept at boiled-off nitrogen over-pressure, 1 mm of cadmium and 20 cm of paraffin (see Figure 3 for a detail of the inner layout of the set-up). DM32 was in operation at the Canfranc Underground Laboratory, in Lab 1 (Figure 1), under a rock overburden equivalent to 675 m of water,

accumulating 4613.6 kg×d of data between May 1993 and December 1994. The energy threshold was at 8 keV[1] and the radioactive background in the low energy region was about 10 cpd/kg/keV [34]. Two scenarios were considered for presenting the results of the experiment: pure spin-independent and pure spin-dependent coupling between DM particles and target nuclei, both in the standard analysis (by comparing the upper limit to the measured background to the expected signal for a given candidate) and the annual modulation analysis. The latter allowed the improvement in about two orders of magnitude of the parameter space ruled-out by the experiment [54,55].

Figure 3. DM32 experiment layout: the three detectors made of 10.7 kg NaI(Tl) inside the lead shielding can be seen. DM32 results can be found in [54,55].

3.2. ANAIS Prototypes

The large expertise acquired by the group using NaI(Tl) scintillators and other techniques applied to the searches for DM pushed forward a new project—ANAIS (Annual modulation with NaI Scintillators). The rationale behind the ANAIS proposal was the positive result of annual modulation of the DAMA experiment [11,12], a few years after DM32 experiment closed operation, which has been puzzling the field for more than twenty years since then, accumulating statistical significance in its observation up to the present [13–15]. A model-independent comparison with the DAMA/LIBRA result was, from the beginning, the goal of the ANAIS project. For that goal, using the same target was mandatory, together with a different experimental approach in order to understand possible systematics. The DM32 experiment concluded that most of the observed radioactive background at low energy could be attributed to ^{210}Pb and ^{40}K in materials close to the crystals or in the crystals themselves. Then, the first ANAIS prototypes [35–37] tried to improve the background by reducing those contributions: replacing the PMTs and the original steel housing of the NaI(Tl) modules, made by BICRON, by more radiopure PMTs and housing. In parallel, the reduction of the energy threshold was targeted by implementing a pulse-shape discrimination procedure to distinguish PMT-origin events from NaI-scintillation events, and the design of a readout by two PMTs per module. A full program of detector upgrading, building three prototypes and carrying out measurements of the corresponding radioactive backgrounds at LSC's Lab 3 allowed us to reduce the background and threshold, but confirmed that the dominant contamination was present in the crystal, showing that an experiment using the available 150 kg NaI(Tl) was not feasible. New radiopure crystal providers had to be searched for (see below). On the other hand, the effect of light guides on the light collection and a better understanding of the light propagation and optical properties of the system was achieved using those prototypes, which allowed the group to gain expertise in the design of good-performing scintillating modules. Figure 4 shows several steps of the procedure developed to couple a NaI(Tl)

crystal to the light guides and the PMTs inside a Teflon housing, everything enclosed in a tight copper box to prevent humidity from harming the crystal (Prototype-II).

Figure 4. Process of installing Prototype-II inside a dry glove box for testing the optical performance. Later, on the Canfranc Underground Laboratory facilities, the measurements allowed to test the radioactive background of the prototype.

For instance, the scintillation of natural quartz was identified as the origin of an anomalous population of events in Prototype III, faster than NaI(Tl) bulk scintillation but slower than typical PMT-origin events [2]. This prototype consisted of a 10.7 kg BICRON crystal installed in a tight OFHC copper housing with two natural quartz optical windows allowing to couple the PMTs in a second step. Such an anomalous population was difficult to disentangle by Pulse Shape Discrimination (PSD), and imposed the careful selection of materials to design a good-performing optical system. Synthetic quartz was studied and no scintillation was found in similar conditions [89], being chosen for the optical windows in subsequent prototypes.

In parallel, as commented before, new providers of radiopure NaI(Tl) crystals were contacted, in particular, Saint Gobain, the DAMA/LIBRA crystals' provider company. A 9.6 kg crystal was acquired as similar as possible to those of DAMA/LIBRA, and it was used to build the Prototype-IV. This crystal was shown to be very good in terms of ^{210}Pb bulk contamination, but not in ^{40}K. Later on, ultra-low-background NaI(Tl) crystals from Saint-Gobain, similar to those of DAMA/LIBRA, were not available, although we contacted the company on several occasions. After exploring how to develop purification protocols in collaboration with other companies, finally Alpha Spectra Inc.[3] (AS), in Grand Junction, Colorado, US, was contacted. They agreed to produce NaI(Tl) following procedures proposed by our group, in particular incorporating a copper housing design with a Mylar window. Size and shape of the modules were also agreed between ANAIS team and AS. Later, this design was used to build modules for other collaborations, as COSINE. The Yale group, which was by the time involved in the DM-ice experiment [47], also acquired AS crystals but decided to change the size and mass of the detectors.

The first two AS modules arrived at LSC in 2012 and were readily characterized [38]. They showed excellent light collection but the background was higher than that of DAMA/LIBRA, mainly because of the high ^{210}Pb contamination of the crystal itself, confirmed by the measurement of the alpha rate. Further collaboration with AS company allowed to reduce significantly this high content in ^{210}Pb in subsequent modules, which however, remains as one of the most relevant contributions to the background budget in the region of interest (ROI), from 1 to 6 keV. Seven additional modules arrived at LSC between 2015 and 2017, completing the ANAIS-112 experiment layout, explained in detail in Section 4.

4. ANAIS-112 Experiment

The ANAIS-112 experiment was the result of a time-extended R&D effort and expertise accumulation in the operation of NaI(Tl) detectors, described in Section 3. The experiment was commissioned during the spring of 2017 and it started the data-taking phase at the hall B of the LSC on 3 August 2017.

4.1. ANAIS-112 Experimental Setup

A full description of ANAIS-112 experimental setup can be found in [67]. It consists of 112.5 kg of NaI(Tl) , distributed in 9 modules, 12.5 kg each and built by Alpha Spectra Inc., arranged in a 3 × 3 configuration. Among the most relevant features of ANAIS-112 modules we can mention a remarkable optical quality, which combined to using high quantum efficiency Hamamatsu photomultipliers (PMTs) results in a very high light collection, at the level of 15 photoelectrons (phe) per keV [66] in all the nine modules. Another interesting feature is a Mylar window in the middle of one of the lateral faces of the detectors, which allows to calibrate the modules with external sources of energies just few keV above the ROI for testing the DAMA/LIBRA result ([1–6] keV).

The ANAIS-112 experiment is installed inside a shielding consisting of an inner layer of 10 cm of archaeological lead and an outer layer of 20 cm of low activity lead. This lead shielding is encased into an anti-radon box, tightly closed and kept under overpressure with radon-free nitrogen gas. The external layer of the shielding (the neutron shielding) consists of 40 cm of a combination of water tanks and polyethylene bricks. An active veto made up of 16 plastic scintillators is placed between the anti-radon box and the neutron shielding, covering the top and sides of the set-up (see Figure 5) allowing to effectively tag the residual muon flux.
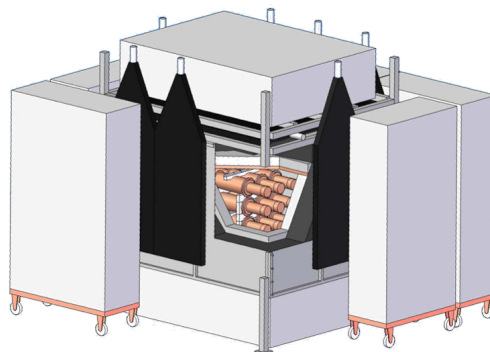

Figure 5. Sketch of the ANAIS-112 setup. It shows the NaI(Tl) modules inside a shielding made of lead, anti-radon box, active muon vetoes, and a neutron shielding made of polyethylene, and water tanks.

The signals from the two PMTs coupled to each module are digitized at 2 GS/s with high resolution. The trigger requires the coincidence of the two PMT trigger signals in a 200 ns window, while the PMT individual trigger is set at the single photo-electron level.

The stability of the experiment is guaranteed by calibrating with ^{109}Cd sources every two weeks. This allows the correction of the small gain drifts. The energy calibration is performed with the ^{109}Cd lines plus the energy depositions at very low energy (3.2 and 0.87 keV) associated with the decay of ^{40}K and ^{22}Na crystal contaminations, respectively, that can be tagged by coincidences with high energy gammas. This procedure allows us to perform a reliable energy calibration of the ANAIS-112 data down to 1 keV, while the identification of the events from ^{22}Na allows us to confirm the highly efficient triggering below 1 keV. We check the stability of the calibration procedure in the ROI by calculating the residuals of the energy associated to the ^{40}K and ^{22}Na peaks with respect to their nominal energy, gathering data every 90 days. We obtain average residuals of 0.01 keV at the 3.2 keV line of ^{40}K and -0.04 keV at the 0.87 keV line of ^{22}Na, the latter below the ROI. The standard deviation is about 0.015 keV for both distributions.

The background in the ROI is dominated by non-bulk scintillation events, so we apply strong filtering protocols based on the pulse shape and light sharing among the two PMTs. The efficiency of the event selection criteria is calculated with scintillation populations (^{109}Cd, ^{40}K, ^{22}Na) and is very close to 100% down to 2 keV, and then decreases steeply to about 15% at 1 keV, where we set the analysis threshold [67].

A careful background model of the ANAIS-112 experiment has been developed over the years, by combining inputs from different analysis techniques: HPGe screening of different materials used in the set-up and in the detector building, measurements in coincidence of the modules before ANAIS-112 commissioning, determination of alpha rate by using PSD, estimates of cosmogenic activation rates in NaI, and so forth [64,65,90,92–94]. A full description of the experiment performance after the first year and a detailed background model can be found in References [67,68]. Considering altogether the nine ANAIS-112 modules the average background in the ROI is 3.6 cpd/kg/keV after three years of data taking [72], while DAMA/LIBRAphase2 background is below 0.80 cpd/kg/keV in the [1–2] keV energy interval, below 0.24 cpd/kg/keV in the [2–3] keV energy interval, and below 0.12 cpd/kg/keV in the [3–4] keV energy interval [14].

4.2. ANAIS-112 Annual Modulation Results

The goal of ANAIS-112 experiment is the testing of the DAMA/LIBRA result on annual modulation and the experiment's design and analysis procedures have been developed having this in mind. This section summarizes the recently published annual modulation results corresponding to three years of data taking.

A blind analysis protocol has been applied: single-hit events in the ROI are kept blinded during the event selection and efficiency calculation from beginning of the data taking, but 10% of the first year's days, randomly chosen, were used for the fine-tuning of the events rejection procedures and background assessment. Up to now, we have carried out three unblindings of our data: at 1.5 years [70], at 2 years [71], and 3 years [72], which correspond to exposures of 157.55, 220.69, and 313.95 kg×y, respectively.

We look for an annual modulation in the same energy regions as DAMA/LIBRA does: [1–6] keV and [2–6] keV with exposures of 1.13 and 2.46 ton×y, respectively. We decided to group the data in 10-day bins, although later, we checked that this choice did not introduce any bias into the result. In a first approach, we worked with the total events rate, adding together the data from the nine modules, and minimize $\chi^2 = \sum_i (n_i - \mu_i)^2 / \sigma_i^2$, where n_i is the number of events in the time bin t_i (corrected by live time and detector efficiency), σ_i is the corresponding Poisson uncertainty, accordingly corrected, and μ_i is the expected number of events at that time bin, that can be written as:

$$\mu_i = [R_0 \phi_{bkg}(t_i) + S_m cos(\omega(t_i - t_0))] M \Delta E \Delta t. \tag{1}$$

Here, R_0 represents the non-modulated rate in the experiment, ϕ_{bkg} is the probability distribution function (PDF) in time of any non-modulated component, S_m is the modulation amplitude, ω is fixed to $2\pi/365\,\mathrm{d} = 0.01721\,\mathrm{rad\,d}^{-1}$, t_0 to $-62.2\,\mathrm{d}$ (time origin has been taken on 3 August and then the cosine maximum is on 2 June), M is the total detector mass, ΔE is the energy interval width, and Δt the time bin width. R_0 is a free parameter, while S_m is either fixed to 0 (for the null hypothesis) or left unconstrained, positive or negative (for the modulation hypothesis).

We considered two different approaches to model the experimental background: (1) following [70,71], the background evolution is approximated to an exponential decay: $\phi_{bkg}(t_i) = 1 + f e^{-t_i/\tau}$, where f and τ are free parameters; (2) following our Monte Carlo background model [68] the background evolution in time is computed and converted into a PDF, so $\phi_{bkg}(t_i) = 1 + f \phi_{bkg}^{MC}(t_i)$. Finally, (3) we considered the nine modules independently, in order to account for systematic effects that could stem from the different backgrounds and efficiencies among detectors. In this third approach, the χ^2 summation was also

performed over detectors and the expected number of events for every time bin t_i and detector d is written as:

$$\mu_{i,d} = [R_{0,d}(1 + f_d\phi_{bkg,d}^{MC}(t_i)) + S_m cos(\omega(t_i - t_0))]M_d\Delta E\Delta t, \qquad (2)$$

where M_d is the mass of every module, $\phi_{bkg,d}^{MC}$ is the PDF sampled from the MC background evolution in time calculated independently for every module, and $R_{0,d}$ and f_d are free parameters for values of d between 0 and 8.

The results of the fit for the three methods and both energy regions are shown in Figures 6–8. Results are also summarized in Table 1.
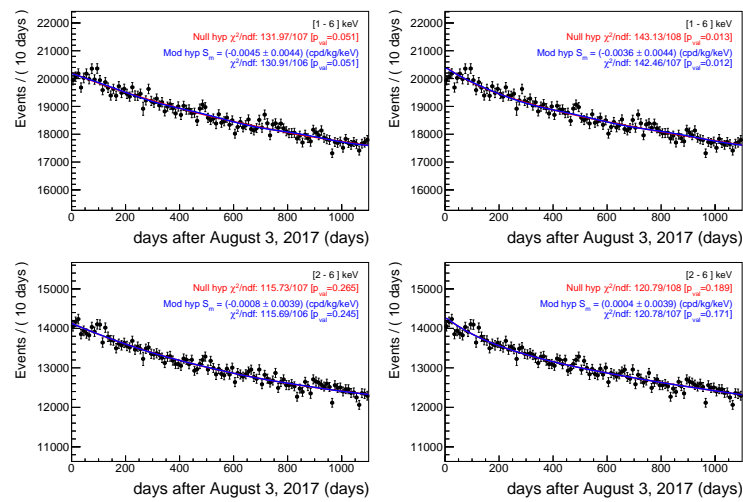

Figure 6. Upper panels: fit results for three years of data in the [1–6] keV energy region in the modulation (blue) and null hypothesis (red) when the background is described by approach (1) (left panel) or approach (2) (right panel). Lower panels: same, but for data in the [2–6] keV energy region.
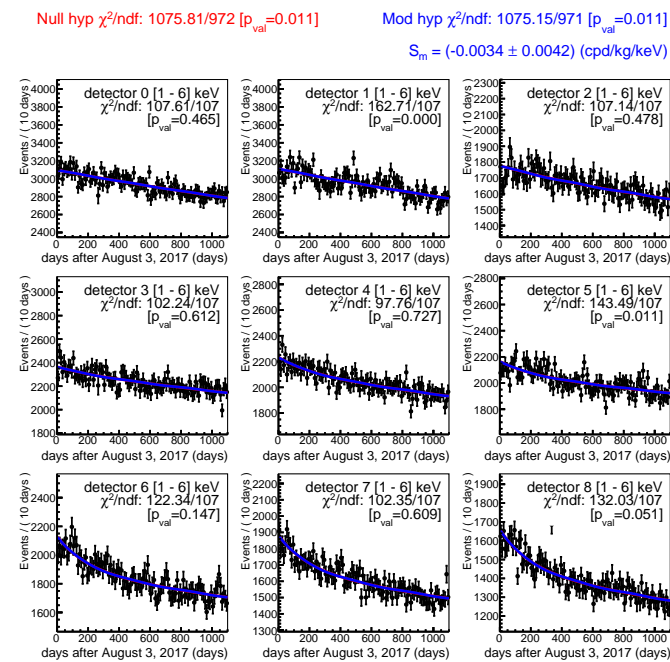

Figure 7. Fit results for three years of data in the [1–6] keV energy region in the modulation (blue) and null hypothesis (red) when the background is described by approach (3).
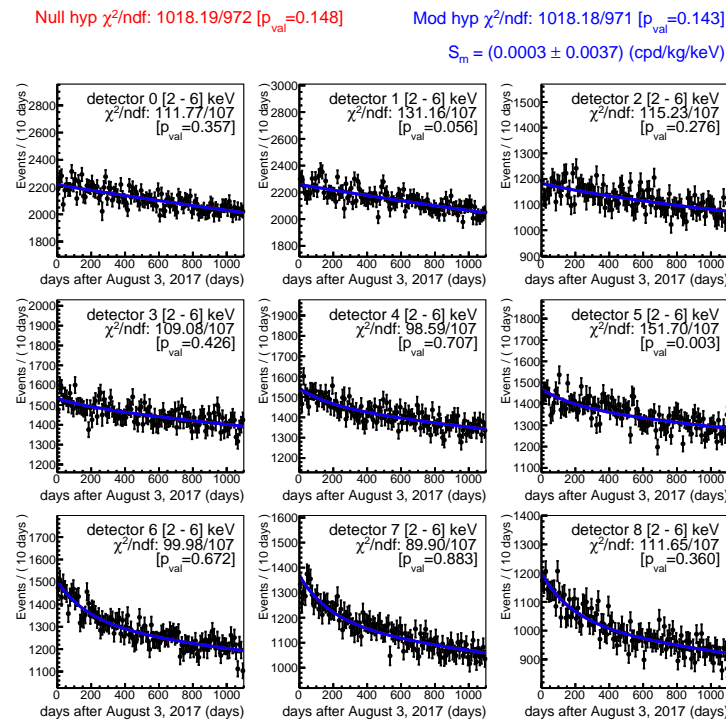
Null hyp χ²/ndf: 1018.19/972 [p$_{val}$=0.148]   Mod hyp χ²/ndf: 1018.18/971 [p$_{val}$=0.143]

S$_m$ = (0.0003 ± 0.0037) (cpd/kg/keV)

Figure 8. Fit results for three years of data in the [2–6] keV energy region in the modulation (blue) and null hypothesis (red) when the background is described by approach (3).

Table 1. Results of the fits for the null and modulation hypothesis with frequency and phase fixed in the ANAIS-112 data corresponding to three years for the three approaches followed for modelling background evolution (see text for more details).

Energy Region	Approach	χ^2/NDF	S_m (cpd/kg/keV)	*p*-Value Mod	*p*-Value Null
[1–6] keV	(1)	132/107	−0.0045 ± 0.0044	0.051	0.051
[1–6] keV	(2)	143.1/108	−0.0036 ± 0.0044	0.012	0.013
[1–6] keV	(3)	1076/972	−0.0034 ± 0.0042	0.011	0.011
[2–6] keV	(1)	115.7/107	−0.0008 ± 0.0039	0.25	0.27
[2–6] keV	(2)	120.8/108	0.0004 ± 0.0039	0.17	0.19
[2–6] keV	(3)	1018/972	0.0003 ± 0.0037	0.14	0.15

In the [2–6] keV region, the null hypothesis in the three approaches describes the data well (*p*-values of 0.27 and 0.19 and 0.15). Smaller *p*-values (0.051, 0.013 and 0.011) are obtained in [1–6] keV region. When we calculate individual *p*-values for every detector, we conclude that only detectors 1 and 5 are responsible of the low agreement with the null hypothesis in the [1–6] keV region (see Figure 7). The anomalous behavior of these two modules could be an indication of noise in the [1–2] keV energy range. The application of machine learning techniques could improve the rejection of non-bulk scintillation events contributing to ANAIS-112 background below 2 keV. We are following this new events selection strategy and the preliminary analysis points at an improvement in the p-values of these two detectors, in particular, which supports the previous hypothesis [41,104].

For the modulation hypothesis, the best fit modulation amplitudes are in all cases compatible with zero at 1σ. The standard deviation $\sigma(S_m)$ is the same for (1) and (2) approaches and slightly lower for the third approach, when detectors are considered independently, as expected following our a priori sensitivity analysis [69]. Therefore, this is the standard deviation chosen to quote the ANAIS-112 annual modulation final result for three years. Figure 9 shows a graphical representation of our best fits (black dots) and sensitivity (colored bands) in comparison with the DAMA/LIBRA results (blue squares).
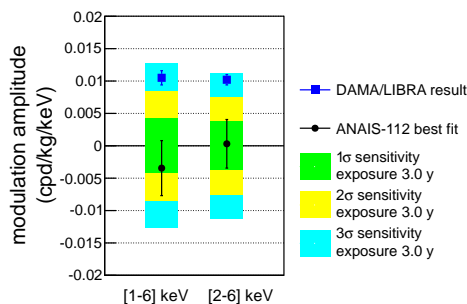
Figure 9. ANAIS-112 result on annual modulation for three years compared to DAMA/LIBRA modulation best fit. Colored bands show the ANAIS-112 estimated sensitivity for such exposure: green at 1σ, yellow at 2σ, and cyan at 3σ.

We have carried out several consistency checks (changing the number of detectors entering into the fit, considering only the first two years or the last two years, or changing the time bin size) and we conclude that we do not have any hint supporting relevant systematical uncertainties in our result. Performing a large set of Monte Carlo pseudo-experiments sampled from the background model, we have checked that the fit is not biased. When we leave the phase t_0 unconstrained, best fits are compatible with no modulation at 2σ, but a bias in the fit in the absence of modulation proportional to $\sigma(S_m)$, the standard deviation of the modulation amplitude, is found to be $\sqrt{\frac{\pi}{2}}\sigma(S_m)$. Correcting the best fits with the calculated bias, our phase-free results are compatible with no modulation in all the approaches and both energy regions at 1σ. We have also performed a frequency analysis, looking for a periodic signal at other frequencies in our data, and the conclusion is that there is no statistically significant modulation in the frequency range searched in the ANAIS-112 data [72].

4.3. ANAIS-112 Sensitivity

In [69] we presented a method to calculate the expected sensitivity to the annual modulation signal according to the measured background, its time evolution and the detector efficiency and live time. Following this method, we have calculated the ANAIS-112 sensitivity projection as a function of the real time starting from 3 August (see Figure 10). The sensitivity is given by the ratio $S_m^{DAMA}/\sigma(S_m)$, which provides, in σ units, the C.L. at which we can test the DAMA/LIBRA result. For three-year exposure, the ANAIS-112 result $\sigma(S_m) = 0.0042$ (0.0037) cpd/kg/keV for [1–6] keV ([2–6] keV) corresponds to 2.5σ (2.7σ) addedsensitivity.

As can be seen in Figure 10, the ANAIS-112 results for 1.5 [70], 2 [71] and 3 years [72] of data-taking fully confirm our previous analysis and this sensitivity projection.
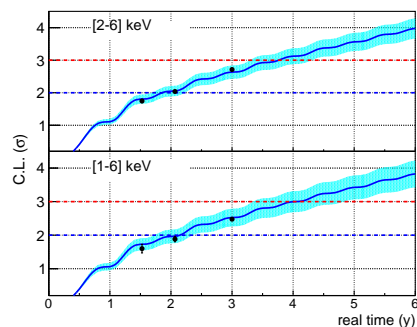

Figure 10. ANAIS-112 sensitivity to the DAMA/LIBRA signal in σ C.L. units (see text) as a function of real time in [1–6] keV (lower panel) and [2–6] keV (upper panel) regions. The black dots are the sensitivities determined experimentally. The blue bands represent the 68% C.L. DAMA/LIBRA uncertainty.

The statistical significance of this result increases as expected and supports our prospects of reaching a sensitivity above 3σ (4σ) in five (six) years of operation. These times can be reduced thanks to the background reduction in the [1–2] keV energy region expected from the improvement in the event selection protocols by the application of machine learning techniques. The preliminary results of this new analysis point to a background reduction of around 25% in this energy region [41].

5. Model-Independent Testing of DAMA/LIBRA beyond 3σ Confidence Level

As commented before, ANAIS-112 will take data until August 2022, when five years of data will be completed and according to our estimates, statistical significance should exceed 3σ (see Figure 10). Up to the date of writing this review, more than 95% of the time has been "live time", useful for analysis. Most of the rest, has been used for calibration of the experiment.

In order to test DAMA/LIBRA beyond 3σ confidence level, several action lines in the frame of ANAIS-112 experiment are under consideration at present. First, we should demonstrate that ANAIS-112 is able to test DAMA/LIBRA in a fully model-independent way (see Section 5.1). Second, extending the data taking for one more year, until August 2023, will allow testing DAMA/LIBRA at the 4σ level with the present experimental ANAIS-112 performance. However, in order to reach 5σ, a much longer period (almost 10 years in total) would be required [41].

ANAIS-112 sensitivity limitation is mostly due to the high background in the ROI, but in particular in the region from 1 to 2 keV. The ANAIS-112 background, as mentioned, is larger than that of DAMA/LIBRA mainly because of the ^{210}Pb and ^{40}K internal contaminations in the crystal. In addition, there is an excess of events with respect to the estimates of the background model in the region [1–2] keV [68], which could be, at least partially, attributed to non-bulk scintillation events leaking from the filtering protocols [67,91]. Reducing these contributions would increase the sensitivity in the [1–6] keV region. This could be achieved in different ways:

- The machine learning analysis based on Boosted Decision Trees (BDTs) under development at present, already commented in Section 4.3, could allow increasing ANAIS-112 sensitivity by reducing the contribution of this anomalous events in the [1–2] keV energy. This analysis could be applied to all the accumulated ANAIS-112 data;
- We are planning to modify the coupling of our PMTs to the NaI(Tl) modules, as the PMTs could be producing this anomalous population of events. ANAIS-112 sensitivity will increase if all the events excess above ANAIS-112 background model estimates disappears. If successful, this strategy would imply to start a new experimental phase, with improved background, and possibly threshold, as the main limitation for threshold reduction is the efficiency of the filtering protocols applied to remove this anomalous population;
- In parallel, we are developing R&D activities addressing the replacement of the PMTs by SiPMs in a longer term, beyond the ANAIS-112 experiment timeline (see Section 5.2);
- In parallel, progress towards a global improvement of the background requires an upgrading in depth of the detectors, which goes beyond the ANAIS-112 experiment timeline. However, it is also the goal of complementary R&D activities (see Section 5.3).

5.1. Towards a Precise Knowledge of the Detector's Response

As stressed in Section 1, a comparison between DM search results obtained with different target nuclei is affected by model-dependencies. Using the same target should allow the removal of these dependencies completely. There is, however, one caveat to this statement—a direct comparison between experiments using the same target is direct only in the case in which the response function of the detectors is fully taken into account in such a comparison of results. This issue is often disregarded, but it is very relevant in the testing of the DAMA/LIBRA result. This is because scintillation is strongly quenched for

energy deposited by nuclear recoils with respect to the same energy deposited by electrons. Experiments are usually calibrated using gammas/electrons and, then, experimental measurements are presented in terms of electron equivalent energy (keV$_{ee}$) and experiments can be directly compared only if energy is deposited by this channel. In the case when DM particles produce nuclear recoils, quenching factors (QF) should be well known to re-calibrate the energy depositions scale.

This is not the case for NaI scintillators. Measurements of QF in the energy region relevant for DM searches, and in particular for the comparison with DAMA/LIBRA in case the signal is attributed to nuclear recoils, do not agree, and in fact are affected by strong discrepancies (QF dependent on the energy versus constant values, for instance) [105–110]. We are collaborating with Duke and Yale researchers to measure more accurately the QF of several crystals in the same conditions in order to determine whether specific crystal properties traduce in a different QF. Preliminary results have been presented recently, but further work in this line is foreseen [111].

In addition, calibrations with neutrons onsite could play a relevant role in DM searches. In the last period of operation of ANAIS-112 we will develop a more complete calibration program using neutron sources.

5.2. R&D Activities to Improve NaI(Tl) Detector Technology: Replacing PMTs by SiPMs

NaI(Tl) detectors are at present state-of-the-art radiation detectors with many applications in very different fields, from environmental monitoring to medical diagnosis. However, their performance for application in rare event searches has progressed slowly. Radiopurity of DAMA/LIBRA crystals has not yet been improved by any other group, for instance, although SABRE, PICOLON and COSINE-200 collaborations are progressing steadily in this direction, growing crystals independently and developing their own protocols [112–114]. As mentioned above, one of the main limitations at present of the application of NaI(Tl) in rare event searches is the light sensor, the PMT. Replacing the PMTs with SiPMs would bring many relevant advantages: lower mass is associated with a lower background contribution from possible radioactive contaminants, quantum efficiency is higher in the wavelength of interest for the readout of NaI(Tl) scintillation, and spurious events originating in the PMTs could be avoided. On the other hand, SiPMs are affected by a much larger dark rate at room temperature, hindering their application for low-energy processes. Very good results have been obtained by cooling the SiPMs in terms of general performance, but in particular in the dark rate. Because of that, efforts to develop a new light readout of NaI(Tl) crystals using SiPMs have started at the University of Zaragoza. The goal is to operate the NaI(Tl) or intrinsic NaI crystal coupled to the SiPMs at very low temperatures—around 100 K—trying to profit from the low dark rate of the SiPM and the light yield increase that could be achieved (at least for the intrinsic NaI) in that temperature range. Working at that temperature is a challenge, but it opens very interesting possibilities—a liquid Argon/Xenon bath could be used simultaneously as active veto shielding and thermal bath, allowing for an additional background rejection strategy. The Canfranc Underground Laboratory disposes of a large tank designed to operate liquid Ar, in the frame of the ArDM [115] and DarkSide [100] projects, that could be used for the testing of a prototype.

5.3. R&D Activities to Improve NaI(Tl) Detector Technology: Growing NaI(Tl) Crystals Underground

Crystal growing for application in rare event searches requires the use of a radiopure powder as input and an extreme ultra-low background environment. Powder purification procedures are usually applied before the growing. Then, it is particularly important to control crucible radiopurity and possible presence of radon in the atmosphere. In addition, re-crystallization and zone-refining procedures can strongly contribute to more radiopure final crystals [116].

However, cosmogenically activated isotopes are being continuously produced while powder and crystals are at the surface, and more significantly if the growing place is at high altitude over sea level, as happens in Grand Junction. It is very interesting to have the possibility of growing crystals underground, shielded from cosmic rays. In the case of NaI(Tl), we have identified that a relevant part of our background in the ROI is associated with tritium, produced cosmogenically [93], but many other isotopes are produced [92]. Although most of them are short-lived, they explain the exponentially decaying background of ANAIS-112 (see Figures 6–8).

We are collaborating on a project to grow radiopure NaI(Tl) crystals underground, at the facilities of the Canfranc Underground Laboratory.

6. Summary and Outlook

Many years of expertise in the operation of NaI(Tl) scintillation detectors at the Canfranc Underground Laboratory support the results of the ANAIS-112 experiment on the model-independent testing of the DAMA/LIBRA observation of an annual modulation with all the expected features corresponding to a DM signal. Five years of data taking will be completed in 2022, allowing for the first time this testing at a 3σ confidence level. No hint of the presence of modulation in ANAIS-112 (three-year exposure analysis) can be reported. The present status of the ANAIS-112 experiment and prospects to continue this research have been presented in this manuscript.

Abbreviations

The following abbreviations are used in this manuscript:

AS	Alpha Spectra Inc.
ANAIS	Annual modulation with NAIs Scintillators
CAPA	Centro de AstroPartículas y física de Altas energías
COSINUS	Cryogenic Observatory for SIgnatures seen in Next-generation Underground Searches
DAMA/LIBRA	DArk MAtter/Large sodium Iodide Bulk for RAre processes
DM	Dark Matter
GIFNA	"Grupo de Investigación en Física Nuclear y Astropartículas" (Research Group in Nuclear and Astroparticle Physics)
LSC	"Laboratorio Subterráneo de Canfranc" (Canfranc Underground Laboratory)
PICOLON	Pure Inorganic Crystal Observatory for LOw-energy Neutr(al)ino
PMT	Photomultiplier Tube
PSD	Pulse Shape Discrimination
ROSEBUD	Rare Objects SEarch with Bolometers UndergrounD
SABRE	Sodium-iodide with Active Background REjection

Notes

1   Unless otherwise stated, throughout this paper, all the energies shown will correspond to electron equivalent energies. This issue is further commented in Section 5.1.

2   These are mainly associated with dark events and Cherenkov emission in the PMT glass being the pulse shape dominated by the Single Electron Response of the PMT.

3   https://alphaspectra.com/ (accessed date: 17 January 2022).

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