Revision Article

Direct Dark Matter Searches Using Sodium Iodide Targets: Status and Prospects

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Abstract

Thallium-Activated Sodium Iodide (NaI(Tl)) scintillators have been widely used for radiation detection since the middle of the XXth century, being applied, for instance, in nuclear medicine, environmental monitoring, nuclear physics, aerial survey, well logging, homeland security, etc. Among other remarkable features, NaI(Tl) offers a very high intrinsic scintillation light yield and ease of growing large-size crystals. On the other hand, the hygroscopic character of the material complicates the manipulation and requires a tight housing of the detector system preventing humidity from reaching the crystal. Energy ranges from a few keV to several MeV are accessible with state-of-the-art technology, using Photomultiplier Tubes (PMTs) for the light readout. These detectors have been successfully applied since the nineties of the XXth century in the direct search for dark matter in the form of hypothetical WIMPs (Weakly Interacting Massive Particles) pervading the galactic halo. WIMPs could explain the galactic dynamics and the 26% of the Universe matter-energy content required to explain the cosmic microwave background radiation anisotropies, and many other cosmological observations, within the standard ΛCDM cosmological model. DAMA/LIBRA experiment, taking data at Gran Sasso National Laboratory (LNGS), in Italy, and using NaI(Tl) detectors, has observed for more than twenty years an annual modulation in the detection rate. This modulation shares all the features expected for the galactic dark matter signal. However, no other experiment has observed any hint supporting the interpretation in terms of dark matter particle interactions of the DAMA/LIBRA result, which seems very difficult to reconcile with the plethora of negative results from different experiments (using different targets and techniques). Only very recently, three-sigma sensitivity to DAMA/LIBRA result is at hand using the same target material, NaI(Tl). This allows us to cancel all the signal dependencies on the particle dark matter model and the dark halo model, enabling a model independent evaluation of that result. In this article, experimental efforts using NaI(Tl) detectors aiming at testing the DAMA/LIBRA signal will be briefly revised, as well as the results released, data-taking status, possible systematics affecting this testing, and sensitivity prospects for the near future. Finally, some R&D efforts toward the development of new experimental approaches using either NaI(Tl) or undoped NaI crystals will be revised in the context of the solving of the DAMA/LIBRA puzzle on annual modulation but also moving forward to other possible applications.

Keywords: Dark Matter, NaI(Tl) scintillators, annual modulation, DAMA/LIBRA result
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1. INTRODUCTION

For almost a century, evidence of the existence of an unknown matter component in the Universe has been accumulating [1]. This matter is required to explain the dynamics of stars and gas clouds in spiral galaxies, galaxies within clusters, cosmic background radiation anisotropies, and a plethora of other astronomical and cosmological observations. The ΛCDM cosmological model, incorporating 68% of the total Universe's mass-energy budget in the form of Dark Energy (DE) and 84% of the matter in the form of Cold Dark Matter (CDM), provides a successful framework able to match the observed Universe properties at the different explored space-time scales, unlike current modified gravity or dynamics theories [2].

The CDM could consist of massive particles with nonbaryonic character, beyond the Standard Model of Particle Physics, weakly coupled to the ordinary matter and stable. This weak coupling could explain that this matter had been considered dark or invisible and it allows their thermal production in the early Universe while opening the door to different detection approaches. Moreover, for a given range of masses, weakly coupled massive particles (WIMPs) would behave as CDM, allowing us to explain the structure formation in the Universe. Many other candidates for this CDM are on scene and finding clues on the DM nature requires complementary approaches, combining searches at accelerators [3], and indirect [4] and direct searches [5, 6, 7]. Despite the large experimental effort and the strong improvement in sensitivity achieved, none of them has succeeded in that goal, but many candidates for this DM component of the Universe have been strongly cornered down by their negative results.

This article focuses on direct DM searches, aiming at measuring the energy directly transferred by a DM particle to a convenient target able to convert this energy into a visible signal. This energy is small and strongly dependent on the DM particle properties (mass and coupling to quarks and/or leptons) and their distribution within our galaxy (DM density and velocity distribution function, both near the Solar System position), which are unknown or affected by large uncertainties. For many DM particle candidates, this energy is expected to be transferred preferentially to the target nuclei by elastic scattering. Because of those unknowns and uncertainties in the DM particle and halo models, the comparison of results among experimental searches with different target nuclei requires choosing some model, and then, the incompatibility among experiments cannot be stated on model-independent grounds.

The low interaction rates expected for these DM particles imply that backgrounds have to be minimized by operating the experiments underground, using radiopure materials in the detector building, shielding conveniently (passively and/or actively) against external backgrounds, and profiting from any background rejection procedure at hand. In spite of all of this, and even achieving a zero radioactive background detector, the neutrino background will be difficult to fight and overcome. This leads us to search for a distinctive feature of the DM signal against other backgrounds. The annual modulation effect in the DM interaction rates plays such a role [8, 9]. It arises from the orbital motion of the Earth around the Sun along the year while the Sun is traveling around the galactic center. The induced variation in the relative velocity of the DM particles with respect to the target nuclei in the detector produces the modulation in the DM interaction rate with a one-year period and maximum at about the 2^{nd} of June, within the standard halo model.

After more than three decades of DM direct searches, most of the experimental results have been compatible with the estimated backgrounds, although some excesses of events have been found [10, 11, 12]. Most of them could finally be identified as backgrounds [13], highlighting that any claim for a detection over the background in this kind of searches requires an extraordinary knowledge of many different background sources, some of them being very specific for the considered detection technique applied. These excesses are easier to find near the energy threshold and whenever a qualitative improvement or the implementation of a new detection technique allows accessing previously unexplored signal realms [14]. On the other hand, there is a positive result, the observation by the DAMA/LIBRA experiment of an annual modulation in the detection rate with a very high statistical significance, accumulating more than two decades of data. This result cannot be explained by any known background, and all the systematic effects proposed to contribute to such a modulated signal have been discarded by the DAMA/LIBRA collaboration [15, 16, 17, 18, 19, 20]. However, although this result is very difficult to reconcile with the incompatible bounds on the DM properties provided by the rest of experiments in most of the considered scenarios [5, 6, 7], it cannot be refuted in a model-independent way, as commented before. Solving this experimental puzzle, which could be the door into new physics, requires the design of model-independent tests. First, using the same target material is fundamental for such model-independent tests. Second, enough sensitivity in the energy region singled out by the DAMA/LIBRA result on annual modulation has to be guaranteed. This requires a good background level and threshold energy, large exposure (product of mass and measurement time), and stability in the operation of the experiment. This is the goal of several experiments, either in data taking or in the R&D phase, such as ANAIS-112, COSINE-100, SABRE-North, SABRE-South, COSINE-200, PICOLON, ASTAROTH, ANAIS+, and COSINUS. Other experiments with different targets have also searched for such an annual modulation in the interaction rate without success [21, 22, 23], but the comparison with the DAMA/LIBRA result, as stressed before, is strongly model-dependent.

Thallium-doped sodium iodide (NaI(Tl)) scintillation detectors are a convenient choice for detecting radiation in many applications, for instance, in nuclear medicine, environmental monitoring, nuclear physics, aerial survey, well logging, homeland security, etc. In particular, they have been applied in direct searches for dark matter for more than thirty years. NaI(Tl) is a high-performing scintillating material: large-size crystals can be grown and the amount of light produced by deposited energy unit is among the highest, at the level of 40000 photons/MeV. The wavelength of the light produced matches perfectly with the peak sensitivity of bialkali PMTs, easing the fabrication of scintillation detectors. In the application to DM searches, NaI has other relevant advantages, as a 100% nonzero spin isotopic content for both ^{127}I and ^{23}Na, which makes this material a remarkable target for spin-dependent interacting DM particle candidates, while the combination of heavy and light nuclei allows a good kinematic matching in a wide range of DM particle masses. In addition, the long-standing DAMA/LIBRA result on annual modulation has kept the interest in this target material alive for DM direct searches.

The radioactive background level achieved by NaI(Tl) detectors has practically not improved in the last twenty years, and it is orders of magnitude above the background level of other experiments, for instance, those using LXe [13]. There are several reasons behind this. On the one hand, raw powder purification and crystal growing technology developed at BICRON/Saint-Gobain at the end of the last century and used to grow DAMA/LIBRA crystals 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 scintillation-based detection 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 that 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 present status and results of those experiments using NaI(Tl) as target material applied to the search for DM; Section 3 focuses on the analysis of some of the systematics jeopardizing the model-independent testing of the DAMA/LIBRA result; Section 4 sets the scene for the future of this technology, looking forward to a definitive testing of the DAMA/LIBRA puzzle but also to other applications.

2. STATUS OF DM SEARCHES USING NaI(Tl) SCINTILLATORS

2.1. DAMA/LIBRA

The DAMA/LIBRA experiment (Large sodium Iodide Bulk for RAre processes) is installed at the LNGS, in Italy, and uses since the start of the data taking in 2003 highly radiopure NaI(Tl) crystal detectors, produced by Saint-Gobain company, of 9.7 kg mass each in a 5×5 matrix configuration. Previously, DAMA/NaI (1995–2002) used only 10 modules of less radiopure NaI(Tl) detectors. The crystals are coupled with two PMTs (one on each side) through 10 cm Suprasil-B light guides. The original PMTs were replaced by High Efficiency (HE) Hamamatsu PMTs in 2010 for the DAMA/LIBRA-phase2, enabling a reduction of the experiment's threshold from 2 to 1 keVee. The crystals, light guides, and PMTs are housed within low-radioactive OFHC (Oxygen-Free High Conductivity) freshly electrolyzed copper shields. The detectors are enclosed in a copper antiradon box continuously flushed with high-purity nitrogen gas. The passive shield is made of copper, low-activity lead, cadmium foils, and polyethylene [24].

First, DAMA/NaI (1995–2002) and later DAMA/LIBRA (since 2003) collaborations have been reporting a positive result in the search for an annual modulation in their detection rates, compatible with the signal expected for DM particles in the standard halo model [15, 16, 17, 18, 19, 20]. DAMA/LIBRA analysis strategy is based on building the residual rate of the single-hit scintillation events in the [2–6] keVee energy region for the full exposure and in the [1–6] keVee for the DAMA/LIBRA-phase2, by subtracting every year the average measured rate in that region. Then, they fit the corresponding residual rates to a function

$$S(t) = S_m \cos\left(\omega\left(t - t_0\right)\right) \tag{1}$$

with free modulation amplitude, S_m, while the period ($T = 2\pi/\omega$) and the phase (t_0) are fixed at 1 year and the 2$^{\text{nd}}$ of June, respectively. This fit results in modulation amplitudes $S_m = 10.3 \pm 0.8$ c/keVee/ton/day at 12.9 σ C.L. in the [2–6] keVee energy region and 10.6 ± 1.1 c/keVee/ton/day at 9.6 σ C.L. in the [1–6] keVee region [20]. However, this procedure is known to be affected by systematics in the case of time-dependent backgrounds, as shown in [25, 26, 27]. A background in the ROI steadily increasing with time would be required in order to explain the DAMA/LIBRA result, which is difficult to justify.

The DAMA/LIBRA collaboration interprets this result as an evidence of the detection of DM, where the signal has all the features expected: it is not observed neither in multiple hit events nor at energies above 6 keVee, it is statistically consistently distributed among the 25 detectors, and it cannot be explained by any background or systematic effect analyzed. Although the signal in the [2–6] keVee region can be interpreted in terms of standard WIMP models, the parameter space singled out by the compatibility with DAMA/LIBRA result for scattering in I and Na (high and low mass regions) has been by orders of magnitude ruled out by many other experiments using different targets and techniques, in both the pure spin-independent (SI) and spin-dependent (SD) WIMP coupling schemes [7]. Although finding compatibility scenarios has become a tough task, a model-independent confirmation is still lacking, and the DAMA/LIBRA result could be the first-ever detection of DM or just some systematics unaccounted for.

Only very recently, experiments using NaI(Tl) detectors have shown enough sensitivity to cast some light on the DAMA/LIBRA puzzle. Both the COSINE-100 and ANAIS-112 experiments use the same target material and detection technique as DAMA/LIBRA, NaI(Tl), but they use a different crystal provider, different growing procedure, raw powder material, and experimental approach. However, DAMA/LIBRA crystals have a significantly lower level of crystal-bulk contamination than COSINE-100 and ANAIS-112 experiments, below 620 μBq/kg for ^{40}K and between 5 and 30 μBq/kg for ^{210}Pb, implying a much lower background in the ROI. DAMA/LIBRA-phase2 background is below 0.80 c/keVee/kg/day in the [1–2] keVee energy interval, below 0.24 c/keVee/kg/day in the [2–3] keVee one, and below 0.12 c/keVee/kg/day in the [3–4] keVee [18].

2.2. COSINE-100

The COSINE-100 experiment was located at the YangYang (Y2L) Laboratory, in the Republic of Korea, and it started the data taking on the 30$^{\text{th}}$ of September, 2016 [28], being very recently decommissioned, in the first half of 2023. It consisted of a total mass of 106 kg of low-background NaI(Tl) crystals produced by Alpha Spectra Inc., set up in a 4×2 matrix configuration. Crystals have different sizes and characteristics. They are hermetically housed in 1.5 mm thick OFC (Oxygen-Free Copper) with two synthetic quartz windows on the sides to couple with two Hamamatsu HE PMTs. All the modules were designed to have windows for energy calibration with external sources, consisting of either a reduced copper thickness of 0.5 mm or a 0.13 mm thick Mylar film, depending on the detector. The modules are housed in an acrylic box, immersed in a 2200 L liquid scintillator tank, which acts as a veto for muon-related events, and other background components, being the ^{40}K emissions from the crystals themselves the most relevant among those contributing in the region of interest (ROI). The tank is surrounded by a shielding consisting of an OFC box, lead, and additional plastic scintillators. As a reference, crystal-bulk contamination in terms of ^{40}K and ^{210}Pb in COSINE-100 crystals is between 580 and 2500 μBq/kg and between 740 and 3200 μBq/kg, respectively.

COSINE-100 experiment searched for excess of events above the background using 59.5 days [28] and 1.7 years [29] of data, exploring different WIMP interaction scenarios, without finding any signal consistent with the DAMA/LIBRA annual modulation result. However, beyond relying on the uncertainties in the background modeling, these results are model-dependent. On the other hand, COSINE-100 presented also an analysis of the annual modulation for the first 1.7 and 3 years of data (the latter for a total exposure of 173 kg×yr), using an effective mass of 61.3 kg [30, 31]. The best fit derived for the modulation amplitude of the single-hit events when the period and phase are fixed, for the three-year exposure, is $S_m = 5.1 \pm 4.7$ c/keVee/ton/day (6.7 ± 4.2 c/keVee/ton/day) for the [2–6] keVee ([1–6] keVee) energy region, consistent with both the null hypothesis and the DAMA/LIBRA's best-fit value [31]. The collaboration is at present commissioning an upgrade of the COSINE-100 setup at the new facilities of the Yemilab Laboratory, in order to continue the data taking with improved performance, while preparing an enlarged experiment, COSINE-200 [32], with more radiopure NaI(Tl) crystals (see Section 2.5).

2.3. ANAIS-112

The ANAIS-112 experiment (Annual Modulation with NaI Scintillators) is the result of a time-extended R&D effort and expertise accumulation in the operation of NaI(Tl) detectors at the University of Zaragoza, in Spain, for about thirty years [33, 34, 35]. The experiment was commissioned during the spring of 2017, and it started the data-taking phase at hall B of the Canfranc Underground Laboratory (LSC) [36], in Spain, on August 3rd, 2017. A full description of the experimental setup can be found in [37]. ANAIS-112 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. ANAIS-112 crystals have a remarkable light collection, higher and more homogeneous than that of the DAMA/LIBRA modules, at the level of 15 photoelectrons/keV [38]. A Mylar window allows us to calibrate the modules with external sources of energies just few keV above the ROI for the testing of the DAMA/LIBRA result. The ANAIS-112 shielding consists of 10 cm of archaeological lead, 20 cm of low-activity lead, an antiradon box (kept under overpressure with radon-free nitrogen gas), and 40 cm of a combination of water tanks and polyethylene bricks. An active veto made up of 16 plastic scintillators covers the top and sides of the set up allowing us to effectively tag the residual muon flux. The signals from the two PMTs coupled with each module are digitized at 2 GS/s with high resolution (14 bits) in a 1260 ns window with a 20% pretrigger region. The trigger requires the coincidence of the two HE Hamamatsu PMT signals in 200 ns, with a threshold at a single photoelectron level in each PMT.

The stability of the experiment is guaranteed by calibrating with ^{109}Cd sources every two weeks. This allows correcting the small gain drifts. The energy calibration is performed with the ^{109}Cd lines and the Br X-rays excited in the source housing, besides the energy depositions at very low energy (3.2 and 0.87 keV) associated with the decay of ^{40}K and ^{22}Na crystal contaminants, which can be tagged by coincidences with high energy gammas in another detector. This procedure allows performing a reliable energy calibration of the ANAIS-112 data down to 1 keV (average residuals of 0.01 keV at 3.2 keV and −0.04 keV at 0.87 keV are obtained when gathering every 90 days coincidence background events), while the identification of the events from ^{22}Na allows confirming the highly efficient triggering below 1 keVee.

The background below 10 keVee is strongly dominated by nonbulk scintillation events, so strong filtering protocols based on the pulse shape and light sharing among the two PMTs have to be applied. The efficiency of the event selection criteria is calculated with the scintillation populations (^{109}Cd, ^{40}K, and ^{22}Na) and is very close to 100% down to 2 keVee but then decreases steeply down to about 15% at 1 keVee, where the analysis threshold is set [37]. However, recently, the development of a new filtering protocol based on machine-learning techniques, in particular boosted decision trees, has allowed increasing significantly the efficiencies for the acceptance of bulk NaI(Tl) scintillation events, while the background was only slightly reduced [39].

A background model of the ANAIS-112 experiment has been developed, by combining inputs from different analysis techniques [40, 41, 42, 43, 44, 45, 46]. Considering altogether the nine ANAIS-112 modules, the average background in the ROI is 3.6 c/keVee/kg/day after three years of data taking [47], considerably higher than that reported by the DAMA/LIBRA experiment. This is because, as commented, crystal-bulk contamination in ANAIS-112 crystals is higher than that of DAMA/LIBRA, while it is very similar to that of COSINE-100 crystals, between 700 and 1330 μBq/kg for ^{40}K and between 700 and 3150 μBq/kg for ^{210}Pb. The ANAIS-112 background in the region from 1 to 2 keVee is above the background model, and understanding the origin of this excess of events remains one of the most relevant systematic issues in the ANAIS-112 analysis. These events could have PMT-origin and leak from the filtering protocol, but an unaccounted-for background source cannot be fully discarded. Work is ongoing in both directions.

Preliminary analyses on annual modulation were released with 1.5 and 2 years [48, 49]. The last published results correspond to three-year exposure [47]. The annual modulation analysis is done in the same energy regions as the DAMA/LIBRA collaboration does in terms of electron equivalent energy: [1–6] keVee and [2–6] keVee, using 10 days time bins and combining the 9-module data. This analysis minimizes $\chi^2 = \sum_i (n_i - \mu_i)^2/\sigma_i^2$, 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, which can be written as follows:

$$\mu_i = \left[R_0 \phi_{bkg}\left(t_i\right) + S_m \cos\left(\omega\left(t_i - t_0\right)\right)\right] M \Delta E \Delta t, \tag{2}$$

where R_0 represents the nonmodulated rate in the experiment, ϕ_{bkg} is the probability distribution function (PDF) in time of any nonmodulated component (including time-independent and time-dependent, but nonmodulated, components), S_m is the modulation amplitude, ω is fixed to $2\pi/365\,\mathrm{d} = 0.01721\,\mathrm{rad\,d^{-1}}$, t_0 is chosen to have the cosine maximum at 2nd June, M is the total detector mass, ΔE is the energy interval width, and Δt is the time bin width. R_0 is a free parameter, while S_m is either fixed to 0 (for the analysis of the null hypothesis) or left unconstrained, positive, or negative (for the modulation hypothesis). Different background time-evolution modeling has been used to derive the results, all of them producing compatible best-fit parameters and p-values. The quoted results in the following are derived from the combined analysis of the rate evolution detector by detector, which follows the same procedure previously explained, but using

$$\mu_{i,d} = \left[R_{0,d}\left(1 + f_d \phi^{MC}_{bkg,d}\left(t_i\right)\right) + S_m \cos\left(\omega\left(t_i - t_0\right)\right)\right] M_d \Delta E \Delta t, \tag{3}$$

where M_d is the mass of every module, $\phi^{MC}_{bkg,d}$ 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.

In the [2–6] keVee region, the null hypothesis describes well the data, while smaller p-values are systematically obtained in the [1–6] keVee region. Analyzing the individual p-values for every detector, it can be observed that detectors 1 and 5 are mostly responsible for the low agreement with the null hypothesis in the [1–6] keVee region. The anomalous behavior of these two modules could be an indication of noise in the [1–2] keVee energy range. This has been checked by applying the new machine-learning filtering protocol, which improves the p-values in those detectors [50].

For an effective exposure of 313.95 kg×yr, the best fit in the [1–6] keVee ([2–6] keVee) energy region results in a modulation amplitude of -3.4 ± 4.2 c/keVee/ton/day (0.3 ± 3.7 c/keVee/ton/day). Therefore, it supports the absence of modulation at 3.3 (2.6) σ, for a sensitivity of 2.5 (2.7) σ [47].

2.4. Model-Independent Comparison

The latest published results [18, 31, 47] of the annual modulation analysis and testing of the DAMA/LIBRA signal are summarized in Figure 1. In the case of ANAIS-112 and COSINE-100 experiments, only the first three years of data have been analyzed and published, although both experiments accumulate by now six-year exposure each. ANAIS-112 sensitivity is also shown in the plot, calculated for the three-year exposure following [51]. In the case of the ANAIS-112 experiment, after the improved sensitivity achieved by the machine-learning-based filtering procedure [39], a sensitivity beyond 4 σ is expected with the available six-year exposure.

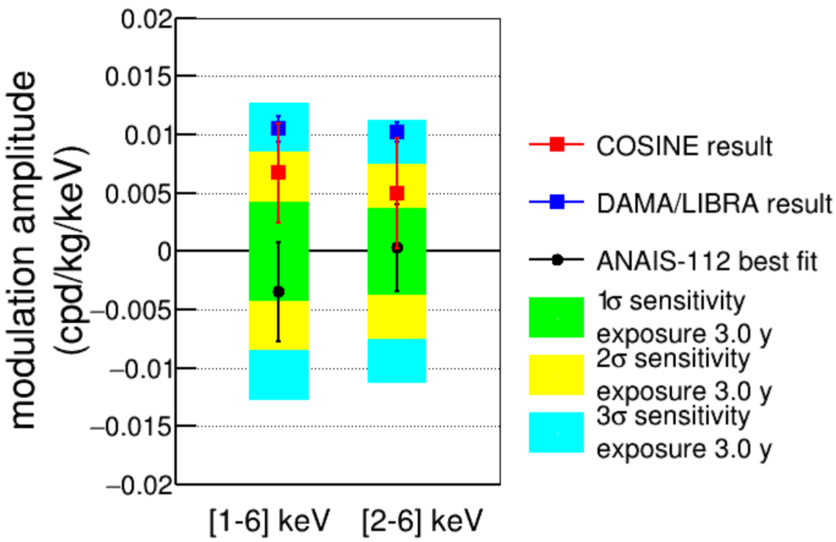

FIGURE 1: Comparison between the results on the annual modulation amplitude best fits from DAMA/LIBRA full published exposure [18] (blue squares), COSINE-100 three-year exposure [31] (red squares), and ANAIS-112 three-year exposure [47] (black dots). The estimated sensitivity of ANAIS-112 for the three-year exposure is shown at different C.L. as colored bands.

The comparison of the three experiments' results relies on a similar response and operation conditions of the detectors. A few comments about this issue are worth remarking on:

(i) ANAIS-112 and COSINE-100 crystals have been manufactured similarly by the same provider, Alpha Spectra Inc., using the same growing technique, similar raw NaI starting powder, thallium content, etc. Therefore, no intrinsic differences are expected to be found in their response, neither to beta/gamma particles nor to neutrons or nuclear recoils. On the other hand, DAMA/LIBRA crystals were made by Saint-Gobain with a proprietary manufacturing protocol, and then, intrinsic differences in the scintillation features between DAMA/LIBRA and ANAIS/COSINE crystals could be present. More on this will be commented in Section 3. In addition, the light collected by ANAIS-112 and COSINE-100 modules is very similar for those modules used in the analysis of the annual modulation. Some of the COSINE-100 modules, larger in size, are not used in the annual modulation analysis because they suffer from a poorer light collection. In the case of DAMA/LIBRA, the light collected per module is smaller and less homogeneous among modules, between 6 and 10 photoelectrons/keV, which could be explained by the light losses in the 10 cm light guides used between the crystal and the PMTs. On the other hand, the effect of these light guides could imply a lower contribution from nonbulk NaI(Tl) scintillation events, originating at the PMTs, for instance.

(ii) Both COSINE-100 and ANAIS-112 have a muon-tagging system, unlike the DAMA/LIBRA experiment. On the other hand, both are installed in laboratories (Y2L and LSC) with a rock overburden inferior to that of LNGS. This could imply a larger contribution from the residual muon flux in the data, which has a seasonal modulation, as confirmed by numerous underground detectors for high-energy muons [52, 53, 54, 55, 56, 57, 58, 59, 60, 61]. Muons have been proposed as a possible origin of the DAMA/LIBRA signal, although the collaboration has always discarded them. It is clear that direct muon interactions should appear at energy regions much above the ROI and at LNGS with a very low rate. However, interactions of the muons or other associated shower particles in other detector components could result in smaller light signals contributing to the ROI, and slow phosphorescence of NaI(Tl) as observed in ANAIS-112 [62] and COSINE-100 is able to produce several triggers with only one primary muon interaction in the detector. Such "muon-tail" events could also appear in the ROI and suffer from time modulation. Both ANAIS-112 and COSINE-100 apply event selection procedures to reject events after a muon is crossing the set up. Several works [63, 64] consider the possibility of radiation-induced delayed light emission leaking through DAMA/LIBRA event selection cuts, which could pass a coincidence trigger and be mistakenly categorized as keV-scale NaI(Tl) scintillation events, prompting again the consideration of seasonal-modulated external sources as a possible explanation for the DAMA/LIBRA signal.

(iii) A different DAQ strategy is followed by the three experiments, using very different digitization signal sampling and operation conditions. ANAIS-112 uses a higher sampling rate and shorter time window than COSINE-100, for instance, and a larger coincidence window for the module trigger between the two PMT signals than DAMA/LIBRA, resulting in a higher trigger efficiency. The sampling rate, ADC-bit resolution, integration window, and trigger efficiency are different for all the experiments. The implications of these differences should be further analyzed.

(iv) Similar HE PMTs are being used for all three experiments. However, very different nonbulk scintillation event rates are observed, being much higher at ANAIS-112, which publishes the lower filtering efficiencies while the light collection in all the ANAIS-112 modules is very high. ANAIS-112 observes an important contribution from asymmetric events difficult to reject by the filtering procedures at hand. Their origin is not clear, but they could be attributed to light produced at the PMTs. In the case of DAMA/LIBRA, this kind of events could be strongly reduced by the light guides present, but it is not clear why the contribution in COSINE-100 is not at the same level as in ANAIS-112 if the origin is in the PMTs. Replacing the PMTs with other light sensors, like SiPMs, could imply a strong boost in the sensitivity of the experiment as will be commented on in Section 4.

2.5. Other Projects

There are other projects in the R&D phase, aiming at improving the radiopurity of the NaI(Tl) crystals but using similar technology than DAMA/LIBRA, COSINE-100, and ANAIS-112 experiments: the detection of the scintillation light of the NaI(Tl) using PMTs.

(i) SABRE (Sodium Iodide with Active Background REjection) collaboration [65, 66, 67, 68] plans to install twin detectors in the Northern and Southern hemispheres, in LNGS (SABRE-North) and Stawell (SABRE-South) laboratories, respectively. The radiopurity level achieved by SABRE's NaI(Tl) crystals is very good in terms of ^{nat}K, 4.3 ± 0.2 ppb,[1] but on the other hand, the contamination achieved in ^{210}Pb is still high: 340 ± 40 μBq/kg [69, 70], and 461 ± 5 μBq/kg ^{210}Pb in the crystal bulk [68].

(ii) The COSINE-200 experiment is the next phase of the COSINE-100 experiment [32], profiting from the results of the R&D on crystal growth aiming at reducing ^{210}Pb and ^{nat}K contamination. The plan is to grow highly radiopure 200 kg of NaI(Tl) detectors, assembled in a low-background environment. They have achieved levels of ^{210}Pb between 10 and 50 μBq/kg and ^{nat}K between 8 and 23 ppb [32].

(iii) The PICOLON (Pure Inorganic Crystal Observatory for LOw-energy Neutr(al)ino) dark matter search project has succeeded in growing crystals with a very low content in ^{210}Pb contamination, below 6 μBq/kg, by using a hybrid purification method, combining recrystallization and ion exchange resins, and ^{nat}K below 20 ppb. They plan to build a 250 kg NaI(Tl) experiment [71, 72, 73, 74].

Other projects in the R&D phase aim at developing a new light readout for the NaI scintillation based on the application of SiPMs, as ANAIS+ [33] and ASTAROTH [75] projects. More information about them will be provided in Section 4.

On the other hand, COSINUS (Cryogenic Observatory for SIgnatures seen in Next-generation Underground Searches) 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 [76, 77, 78, 79]. In this case, important differences among the experimental outputs with respect to the other projects are to be expected. In particular, this bolometric hybrid detection approach, besides the background discrimination capability, allows for a very low threshold in energy and the exploration of the nuclear recoil energy directly, without quenching factor corrections, which is one of the possible systematics affecting the comparison among experiments, as it will be further commented in Section 3.

[1] 1 ppb of natural potassium is equivalent to 30.96 μBq/kg of ^{40}K.

3. NaI(Tl) RESPONSE IN THE ROI FOR DM SEARCHES: LIGHT YIELD NONPROPORTIONALITY AND SCINTILLATION QUENCHING

As stressed before, comparison among experiments carried out with the same target nuclei can be done in a model-independent way, as far as all the dependencies on the halo properties and DM particle interaction model should contribute exactly in the same way. However, the response of the detector should have been taken into account appropriately before this comparison was done. This section focuses on some issues on the response of NaI(Tl) detectors that could affect the comparison among the signals observed by DAMA/LIBRA and the other experiments. All of these points should be conveniently addressed by every experiment.

3.1. NaI(Tl) Nonproportionality in the Light Yield

NaI(Tl) scintillators are well-known nonproportional scintillators in the ROI for dark matter searches. The light yield of NaI(Tl) changes at a few percent level in the range of up to 20 keV [80, 81, 82, 83, 84, 85, 86, 87].

This behavior supposes a relevant systematic question to be faced: how to properly calibrate the ROI of the experiment. Different approaches are followed by the experiments, but it is important to remark that the nonproportional behavior in NaI(Tl) is difficult to model. In the ANAIS-112 experiment, the option followed consists in linearizing the detector behavior in a region as close as possible to the ROI, which, in the end, eases the data treatment and reduces systematics. ANAIS-112 calibration procedure has been described in Section 2.3. DAMA/LIBRA experiment uses for periodical calibration an ^{241}Am source every $\approx$10 days. A good linearity is observed in the data of DAMA/LIBRA-phase1 (established with the 59.5 keV line from ^{241}Am and checked with the 3.2 keV line from the ^{40}K internal contamination tagged by coincidences between modules). However, in DAMA/LIBRA-phase2, slight nonlinearity is observed, resulting in a shift of 0.2 keV at the software energy threshold and being negligible above 15 keV. This nonlinearity is introduced in the analysis following [18]. On the other hand, the COSINE-100 experiment is calibrated using external sources and emissions from internal contaminants in the crystals. Below 70 keV, they observed nonlinear light response and they account for it via an empirical model based on the data provided by the low-energy internal decays (0.9 keV from ^{22}Na, 3.2 keV from ^{40}K, 25.5 keV from ^{109}Cd, 30.5 from ^{121}Te, 49 keV from ^{210}Pb, and 67.8 keV from ^{125}I). More details on the method and the nonlinear detector response function considered can be found in [88].

The different calibration procedures followed could introduce some systematic effect in the definition of the analysis region considered for the annual modulation search. However, we do not expect this effect to be much relevant, as far as backgrounds in the ROI are present, enabling a good control of the calibration residuals for the known energy peak from ^{40}K, at 3.2 keV, for instance.

3.2. Scintillation Quenching Factor for Nuclear Recoils

The scintillation quenching factors (QFs) for sodium and iodine recoils in NaI(Tl) have to be known accurately to compare results from NaI(Tl) experiments with those from other targets if the signal searched for is attributed to nuclear recoils, but also among themselves if QFs are not an intrinsic property of the NaI(Tl) material. As commented in Section 1, nuclear recoils are expected to be produced by the DM interactions in the most standard scenarios. The QFs allow conversion from the nuclear recoil energy scale to the electron equivalent energy scale (usually referred to as visible energy scale as far as the experiments are calibrated with electron/gamma sources).

The measurement of the sodium and iodine scintillation QFs in NaI(Tl) is then a crucial issue for direct DM searches, and it has been faced since the 1990s of the past century. These QFs are defined as the light signal produced by a nuclear recoil with respect to the light signal an electron recoil releasing the same energy would produce. Lacking a theoretical framework to explain this scintillation quenching, experimental measurements are a must.

The principle followed in most of the measurements carried out is to induce nuclear recoils using quasi-monoenergetic neutrons from a beam, pulsed if possible. These neutrons will elastically scatter in the crystal at a known angle, and the scattered neutron can be detected in an array of neutron detectors (backing detectors) covering a significant range of scattering angles. The coincidence of the signal between one of those detectors and the crystal under analysis allows us to identify the energy deposited by the neutron in the scattering (E_{NR}), while the corresponding visible energy (E_{ee}) is determined with the light signal produced in the crystal, conveniently calibrated using electron recoil events. Then, the QF can be calculated as QF $= E_{ee}/E_{NR}$.

The results of some sodium (iodine) QF measurements are shown in Figure 2 [89, 90, 91, 92, 93, 94, 95, 96, 97] (Figure 3 [89, 90, 92, 93, 94, 96, 97]). It can be observed that there is a high dispersion among the measurements, although the most recent of them for the sodium QF agree with a decrease of this factor at low nuclear recoil energies. This dispersion could be due to different systematics affecting each experiment since the experimental set ups and the data analysis are different, but it is also possible that QFs are different for crystals with different properties, such as impurities, Tl content, etc.

Two different scenarios are at present consistent with the available experimental data on sodium and iodine QFs in NaI(Tl):

(i) QF is an intrinsic property of the NaI(Tl) material. Differences in the measurements can be attributed to systematic effects not properly accounted for. In this case, different NaI(Tl) experiments could be compared without problems among them, both in nuclear recoil energies or in electron equivalent energies, independently of knowing or not accurately the value of the QF. However, to compare NaI(Tl) experiments' results with those from other targets in terms of nuclear recoil energy, a better modeling or understanding of the QF should be achieved.

(ii) QF in NaI(Tl) crystals is not an intrinsic property, and it depends on the specific features of each crystal. In this case, comparison among NaI(Tl) experiments for the DAMA/LIBRA testing will require accurate determination of the QF values, independently, for each experiment.

At present, there is not enough piece of evidence to prefer any of both scenarios. Further efforts to understand these QFs are required, because it is clear that the uncertainties in the knowledge of the QFs in NaI(Tl) detectors are an important systematic effect, limiting at present the testing of the DAMA/LIBRA result.

To measure the QF of the NaI(Tl) crystals, a collaborative effort between members of COSINE-100, COHERENT, and ANAIS-112 experiments started in 2018. The crystal dependence of the QF and the systematics affecting the measurements were evaluated by measuring different crystals in the same set up at the Advanced Neutron Calibration Facility, at the Triangle Universities Nuclear Laboratory (TUNL), Durham (US), and applying the same analysis protocol.

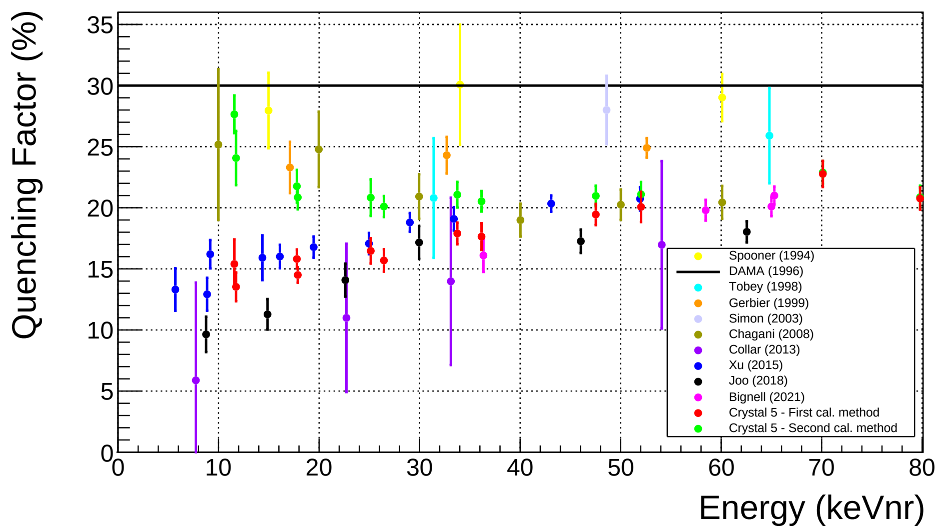

FIGURE 2: Sodium QF results from different experimental measurements [89, 90, 91, 92, 93, 94, 95, 96, 97]. In particular, results for crystal 5, taken from one of the ingots used to build ANAIS-112 crystals, are shown for two different energy calibration schemes ((1) proportional calibration using the ^{127}I inelastic peak as the only reference, and (2) linear calibration using the data from an external ^{133}Ba source). Figure taken from [97].

This measurement has incorporated some specific design features to fight systematics. Small crystals with different sizes and characteristics have been used in the measurements. The same PMT has been coupled with all of them in a similar set up. Two batches of measurements (in August and October 2018) were carried out, with slightly different neutron energy and positioning of the neutron detectors to determine the scattering angles and, then, the nuclear recoil energies. A possible gain drift of the detector system has been corrected by using the information from the ^{127}I inelastic peak along the measurements, while periodic calibration runs with a ^{133}Ba source were also carried out. To avoid threshold effects, the trigger was based on the backing detectors' signal, without requiring a coincident signal in the NaI(Tl) crystal. The NaI(Tl) energy deposition is built by integrating the corresponding crystal waveform in a window fixed by the time of flight of the neutron between both detectors. The energy of the initial neutrons is determined with dedicated time of flight measurements, using a neutron detector in different positions along the beam axis. In addition, a full simulation of the setup and the measurements has allowed us to build PDFs for the nuclear recoil spectra in the NaI(Tl) for coincident signals with every backing detector. These PDFs incorporate all the information corresponding to the dispersion in the neutron energy distribution, the angular size of the neutron beam, and all the sizes of the detectors involved in the setup. Moreover, the uncertainties in the detector's positions can be taken into account to derive systematic uncertainties in the QF results. These PDFs for both sodium and iodine recoils are used for fitting to the experimental measurements, taking also into account the background contribution.

The results of these measurements for one of the crystals measured (crystal 5) are shown in Figures 2 and 3 [96, 97] and they will be published soon. A thorough analysis of systematics has been carried out, some of them being incorporated in the error bars shown in Figures 2 and 3. The most relevant systematics are the modelling of the detector energy resolution in the ROI and the energy calibration. The former is included in the error bars shown in the figures, but not the latter, for which two different energy calibration schemes have been considered: (1) a proportional calibration using the inelastic iodine peak, and (2) a linear calibration with data from the Ba-source in the ROI. Both provide incompatible results for the sodium quenching factors, and then, both results are shown, independently, in Figure 2 for crystal 5, which was taken from the same ingot of some of the ANAIS-112 modules. However, all five measured crystals showed compatible results within the two different calibration schemes. The proportional calibration reproduces the reduction in the sodium QF value observed by many of the most recent measurements at low energies, while the linear calibration is compatible with a constant quenching factor (being the average result $QF_{Na} = (21.2 \pm 0.8)\%$). Finding and quantifying this systematic effect is a very relevant result of this work, and further effort is required to better understand the response of these detectors to nuclear recoils. NaI(Tl) is known as a nonproportional detector in that range of energies, which

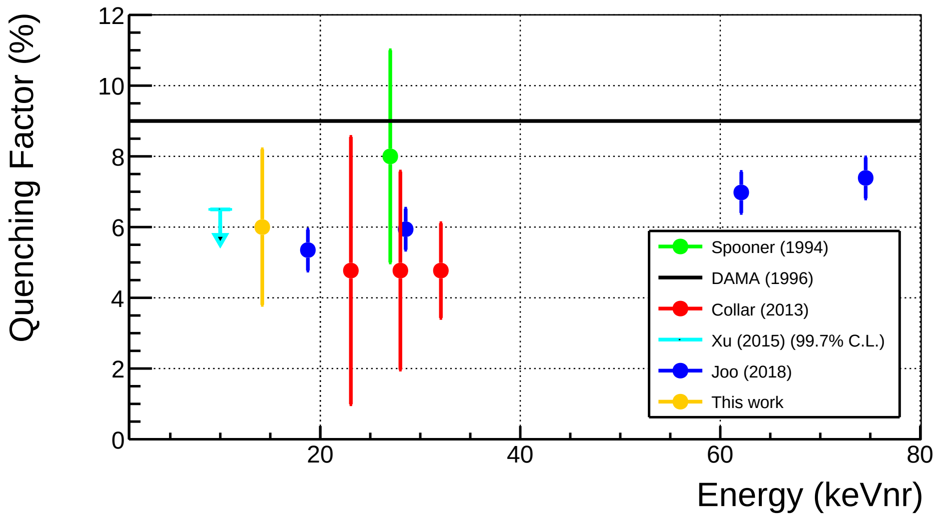

FIGURE 3: Iodine QF results from different experimental measurements [89, 90, 92, 93, 94, 96, 97]. Figure taken from [97].

makes some preference for the nonproportional calibration clear. However, the energy range of interest for the QF analysis requires the extrapolation of such a calibration at lower energies, and moreover, surface or local effects in the interaction of external gamma or X-ray sources could produce some miscalibration or energy artifacts at such low energies. This makes that, at this point, the measurements results are affected by systematics and further work is required for better modeling the behavior of the scintillation QF in NaI(Tl) detectors. One of the lines of research within the ANAIS project is the development of a neutron calibration program of the ANAIS-112 crystals *onsite* which could provide more information on the response function of NaI(Tl) detectors to nuclear recoils (see below for more details).

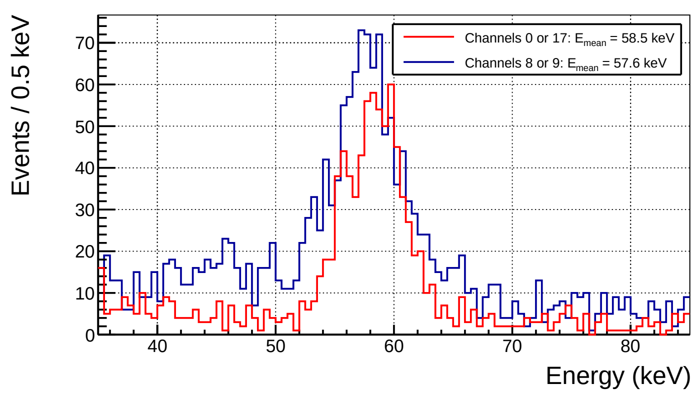

FIGURE 4: Shift on the position of the ^{127}I inelastic peak for reference channels (blue line) with negligible contribution from the nuclear recoil energy and those channels with the highest possible nuclear recoil energy added (red line) for crystal 3 in the measurements at TUNL. Figure taken from [97].

In the case of the iodine quenching factor, the method previously explained for sodium recoils cannot be applied as the recoil peaks are not disentangled from the background. In this case, a different procedure has been followed by analyzing the position of the inelastic peak from ^{127}I, which consists of an energy deposition of 57.6 keV from a gamma, added to the iodine recoil energy. A shift in the peak position is observed, as shown in Figure 4, which allows us to derive $QF_I = (6.0 \pm 2.2)\%$ by averaging results for crystals 2 and 3, while the other three crystals' data only allowed us to set upper limits.

Recently, using the same TUNL facilities a very interesting and complementary program is ongoing to measure the QF for crystals having different thallium dopant content, varying from 0.1 to 0.9% in the initial powder. Preliminary results have been released [98] yielding a value in the range of 0.2 for the different crystals tested assuming a multilinear calibration scheme with an external source similar to that followed in [96, 97].

DAMA/LIBRA presents the results in terms of electron equivalent energy, and then, the model-independent testing done up to this moment by ANAIS-112 and COSINE-100 experiments is only valid in the case the DM particle interacts through transfers of energy to the electrons. COSINE-100 already considered the effect of the different QFs but in a model-dependent approach [29]. Trying to face the incorporation of the uncertainties in the QF's knowledge in a model-independent way, we remark that in the assumption of constant QF (independent of the energy), considering the values 0.3 for DAMA/LIBRA and 0.2 for ANAIS-112 in the case of sodium recoils, and 0.09 and 0.06, respectively, for iodine recoils, the same ratio is obtained for both QFs between DAMA/LIBRA and ANAIS-112: 3/2. This implies that the [2–6] keVee range in DAMA/LIBRA is converted into the [1.3–4] keVee range in ANAIS-112, if we want to integrate the equivalent region in terms of nuclear recoil energies. Equivalently, the [1–6] keVee region converts into [0.7–4] keVee, which is presently below the analysis threshold of ANAIS-112.

Because the uncertainties are still very large, DAMA/LIBRA collaboration should measure again the quenching factor of their crystals, as far as the method followed according to [90] directly assumes a constant QF and it is affected by more systematics than the approach followed by most of the other measurements. ANAIS-112 is reanalyzing also the annual modulation in the [1.3–4] keVee energy region, as a first step to try to evaluate the systematic effect of the QFs in the model-independent testing of the DAMA/LIBRA result.

Within the ANAIS project, there is a neutron calibration program *onsite* of ANAIS-112 detectors under development. A low activity ^{252}Cf neutron source has been used for such a calibration, placed outside the shielding, to produce low-energy nuclear recoils. The large size of the ANAIS-112 crystals makes this measurement be dominated by multiple scattering. The full ANAIS-112 set up has been MC modeled with the GEANT4 package [99] to reproduce the detector response to nuclear recoils. Each energy deposition is corrected by the QF corresponding to the precise energy and particle type, and then, different QF models can be explored and compared to the data. This work is still ongoing, being complementary to the measurements carried out under neutron beam irradiation, and it has shown to be sensitive to different QF models both for Na and I nuclear recoils in NaI(Tl). Preliminary results have been recently released [100].

4. CHALLENGES FOR NaI-BASED SCINTILLATOR DETECTORS

NaI(Tl) detectors can be considered state-of-the-art radiation detectors. However, their performance for the application in rare event searches has progressed slowly.

First, the radiopurity of the DAMA/LIBRA crystals has not yet been improved by any other experiment, although SABRE, PICOLON, and COSINE-200 collaborations are progressing steadily in this direction, and this goal seems to come closer. They have independently designed growth protocols [69, 70, 74]. Crystal growing for application in rare event searches requires using a radiopure powder as input and an extremely ultra-low-background environment along the whole process. 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, recrystallization and zone-refining procedures can strongly contribute to more radiopure final crystals [70]. At present, the ^{40}K activity seems to be under control, but on the other hand, reducing the ^{210}Pb contamination in the large size crystals has shown to be a difficult task.

However, isotopes cosmogenically activated are being continuously produced while powder and crystals are at the surface, more significantly if the growing place is at a high altitude over sea level, as it happens in Grand Junction, Colorado, USA, where the Alpha Spectra company is located. It is very interesting to have the possibility of growing crystals underground, shielded from cosmic rays. In the case of NaI(Tl), a relevant part of the ANAIS background in the ROI is associated with tritium, produced cosmogenically [44], but many other isotopes are also produced [42]. Although most of them are short-lived, they explain the exponentially decaying background observed by COSINE-100 and ANAIS-112 experiments [49, 31]. There are projects to grow radiopure NaI(Tl) crystals underground, which could produce highly radiopure crystals, without such cosmogenic contributions.

Second, one of the main limitations at present for the application of NaI(Tl) in rare event searches is the light sensor typically used, the PMT. Replacing the PMTs with SiPMs would bring many relevant advantages: lower mass is associated with lower background contribution from possible radioactive contaminants, the quantum efficiency is either similar, or even higher, in the wavelength of interest for the readout of NaI(Tl) scintillation, and spurious events originated 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 dark rate. Efforts to develop new light readout of NaI(Tl)/NaI crystals using SiPMs have started recently, including the ASTAROTH project in Italy [75], the ANAIS+ project at the University of Zaragoza, and also dedicated efforts within the COSINE collaboration [101]. ANAIS+ is a project led by the ANAIS team, with the goal of developing a prototype of NaI(Tl) or intrinsic NaI crystal coupled to SiPMs at temperatures around 100K, 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 [102]. This experimental approach 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. First prototypes are under testing, and within the next years, a full program of performance assessment will be carried out, considering tests underground and inside liquid argon.

COSINUS experiment profits from the simultaneous measurement of the light and heat produced in a particle interaction within the sodium iodide target when operated in the millikelvin temperature range. Because of the properties of this material, having a low melting temperature and high hygroscopicity, COSINUS collaboration has developed a novel detection strategy for the heat channel using the "remoTES" design: a Transition Edge Sensor (TES) acting as a thermometer is attached to an external wafer crystal that is thermally coupled to the sodium iodide crystal [78, 79]. For the light channel, a more conventional silicon bolometer is used. Among the advantages of using this technique applied to DM searches is to highlight, first, the full sensitivity to nuclear recoil energy depositions in the heat channel, which removes one of the main systematic effects in the comparison between the DAMA/LIBRA results and those from other experiments; the very low-energy threshold at reach and the background rejection capability this hybrid detection offers. On the other hand, reaching a large exposure can be complicated. In any case, many relevant outputs can be expected from these new experimental approaches in the future.

5. SUMMARY AND OUTLOOK

NaI(Tl) detectors are very interesting for many applications in the field of rare event searches, and they could play a complementary role to detectors using different target nuclei and detection techniques applied in the DM searches, where a multitarget and multitechnique approach could allow us to cope with the many unknowns and uncertainties in the signal searched for. However, the required improvement in the detector performance imposes the introduction of some innovation in the NaI(Tl) readout, profiting from the new technologies in development, in particular SiPMs as light sensors, and the development of specific radiopurity techniques in the growth and manufacturing processes. Working with the crystals at low temperature, in both the approaches followed by COSINUS experiment (in the mK range) and others as ANAIS+ (around 100K), could bring a strong increase in sensitivity, granting access to unexplored energy regions below 1 keV, which would make these detectors competitive targets for dark matter searches in the case of light WIMPs with SD coupling, but also for neutrino coherent scattering, for instance.

Moreover, better strategies progressing toward open-science scenarios could make it easier to settle down puzzles as DAMA/LIBRA in the future. Sharing data in convenient formats is one of the best ways to achieve this. ANAIS-112 offers the release of all the data, accompanied by convenient scripts for reproducing all the published results. This has been already done with the three-year exposure results [47], which are available on the website of the ORIGINS Excellence Cluster: `https://www.origins-cluster.de/odsl/dark-matter-data-center/available-datasets/anais`. Subsequent data releases are in preparation.

CONFLICTS OF INTEREST

The author declares that there are no conflicts of interest regarding the publication of this paper.

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