Scintillator-based detectors for dark matter searches I

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Abstract. One of the widely adapted experimental techniques for the direct search for dark matter is the use of inorganic crystal scintillators. Several experimental groups have carried out direct dark matter search experiments using NaI(Tl), CsI(Tl) and CaF₂(Eu) crystals. The history, status and critical issues regarding these crystal scintillators as dark matter detectors are reviewed in this paper, and the prospects of performing future dark matter searches with these detectors are discussed.

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1. Introduction

The existence of dark matter offers a good explanation for various astronomical observations that cannot be explained well by the elementary particles and fundamental interactions among them described by the Standard Model (SM) of particle physics. The observed rotational velocity curve of our galaxy indicates the existence of dark matter halo in our galaxy. Weakly Interacting Massive Particles (WIMPs), originally introduced as a heavy neutrino [1], are good candidates for particle dark matter. The neutralino, often regarded as the lightest supersymmetric particle (LSP) in supersymmetric (SUSY) models, is a well-motivated WIMP candidate if R-parity is conserved [2]. The axion [3], with a mass of 1 keV or less, is also a good candidate for particle dark matter. It was suggested that an elastic WIMP interaction with an ordinary nucleus may be detected by measuring the small recoil energy of the scattered nucleus [4]. Due to the revolution of the Earth around the Sun, the WIMP velocity relative to a detector fixed on Earth varies with an annual cycle, therefore an annual modulation of event rates in a given energy window is expected [5, 6]. The annual modulation provides an additional handle to confirm the WIMP signature. To measure the tiny recoil energy, lower than 100 keV, of a nucleus from a WIMP interaction, several experimental techniques have been developed over many years. One of the well-established and therefore widely adopted experimental techniques in the beginning of WIMP search experiments was the use of inorganic scintillation crystals.

Inorganic scintillation crystals have been developed for several decades and have been used in many experiments, from nuclear gamma ray spectroscopy to collider experiments at high energies. They have also been used for industrial applications, ranging from medical imaging to x-ray inspection of large vehicles. In high-energy physics, a large number of crystals have been used in various accelerator-based experiments. For example, about 40 tons of CsI(Tl) crystals are used in the BELLE experiment as an electromagnetic calorimeter [7]. Similar amounts are used by the BABAR [8] and BES detectors [9]. Thanks to this demand during the last couple of decades, techniques to grow large amounts of high-quality and large-sized scintillation crystals have been developed to advance the state of the art.
Table 1. Properties of target materials.

| Atom | Z  | A   | J   | ⟨S_p⟩ | ⟨S_n⟩ | Reference |
|------|----|-----|-----|-------|-------|-----------|
| Na   | 11 | 23  | 3/2 | 0.248 | 0.020 | [13]      |
| Cs   | 55 | 133 | 7/2 | −0.370| 0.003 | [14]      |
| I    | 53 | 127 | 5/2 | 0.309 | 0.075 | [15]      |
| F    | 9  | 19  | 1/2 | 0.441 | −0.109| [15]      |

Since these crystals have very good scintillation properties and high light yields, they are suitable for low-energy measurements as well. However, the crystals used for the accelerator experiments were not optimized for low-energy measurement due to the nature of the experiments, namely high-energy particle detection. For example, BELLE crystals are used to measure gamma rays with energies above 50 MeV, so no attention was paid to the internal background of a few MeV or less. In addition, the time window for an event is well defined by the time structure of particle beams in an accelerator experiment, so the randomly occurring low-energy internal radiation does not contribute to the signal. Although a huge amount of crystals has been produced worldwide, most of these are not suitable for dark matter search experiments due to the high internal background. Special care must be given to the whole process, including material preparation, crystal growth and the assembly of the detection system.

Because the nuclear recoil energy to be measured is typically less than 100 keV, it is necessary that the target should be an active detector that can measure such a small energy deposition. Therefore, crystal scintillators made of target elements that have large cross sections for WIMP interaction are preferred. WIMP interactions can be spin independent (SI) or spin dependent (SD), depending on the nature of WIMP particle candidates. The cross section for SI interactions is proportional to the atomic mass squared, \( A^2 \), while the SD cross section is proportional to \( \frac{J(J+1)}{2} \langle S_p, n \rangle \), where \( J \) is the total angular momentum and \( \langle S_p, n \rangle \) is the spin expectation value of a nucleus. Therefore, nuclei with odd number of protons or odd number of neutrons are sensitive to the SD interaction. Both NaI(Tl) and CsI(Tl) crystals have been used for dark matter searches. As a target for WIMP interactions, Na, Cs and I have good sensitivity to the SD interactions of WIMPs as well as to the SI interactions. The properties of these target elements are given in table 1. In addition, it is fairly easy to acquire a large quantity of crystals at a reasonable price. This is an attractive feature as one needs about 100 kg or more to perform an annual modulation search with good sensitivity.

NaI(Tl) crystals have been used by several groups, such as DAMA, ANAIS, NAIAD and ELEGANT V. The DAMA experiment reported an annual modulation of event rates at low energies. However, no positive identification of nuclear recoil was attempted for those events. Nevertheless, this modulation may be interpreted as evidence of WIMP interactions in their detector. However, the indicated signal region with the commonly quoted WIMP model is not consistent with several other experiments using techniques that identify the nuclear recoil signals, such as CDMS and XENON. CsI(Tl) crystals have not been used for dark matter searches until recently, mainly due to the contamination of radioactive isotopes of Cs. KIMS recently developed radio-pure CsI(Tl) crystals and started to use them for the search for dark matter.

The CaF\(_2\)(Eu) crystal is also a good dark matter search detector because of its high sensitivity to the SD WIMP–nucleus interaction, thanks to \(^{19}\text{F}\), which has a large spin expectation.
value, as shown in table 1. Searches for dark matter with CaF$_2$(Eu) have been carried out by BPRS, Osaka and Tokyo groups. This crystal is also one of the good candidates for a $^{48}$Ca double beta decay search. Among other crystals that contain $^{19}$F, LiF was used for WIMP search by means of the bolometer technique by the Tokyo group [11]. Another good example of using a scintillation crystal for a bolometric detector is CaMoO$_4$. CRESST has used the CaWO$_4$ crystal to read out scintillation light and heat signals simultaneously, which allowed a good rejection of gamma background [12].

In this paper, we review the history, status and critical issues regarding crystal scintillator-based dark matter searches, especially on non-bolometric detectors, such as NaI(Tl), CsI(Tl) and CaF$_2$ crystals. The prospects of performing future dark matter searches with these detectors are also discussed.

2. Characteristics of crystals

Some alkali halide crystals with a small concentration of impurity atoms as an activator, such as NaI(Tl) and CsI(Tl), are the most luminescent among the scintillators currently available in large quantities. The techniques to grow large-sized high-quality crystals of these types are very well established. NaI(Tl) crystals are highly hygroscopic and one needs to encapsulate them to protect them from moisture. CsI(Tl) crystals are slightly hygroscopic and encapsulation is not usually necessary, but keeping the crystals in a dry condition is desirable for long-term stability. CaF$_2$(Eu) is not hygroscopic so no special care is necessary.

2.1. Scintillation properties

Pure NaI and CsI crystals without doping are not very efficient scintillators at room temperature, although they can be good scintillators at low temperature. By adding a small amount of impurity atoms, such as Tl, Na and CO$_3$ as luminescent centers, they can be made into efficient scintillators at room temperature. It is known that a doping concentration of Tl higher than 0.07 mol% does not improve the light yield since the self-absorption of scintillation light increases [10]. Although the decay times of scintillation light of pure CsI and NaI crystals are within a few tens of ns, these become longer for doped crystals. For example, CsI(Tl) has more than two light emission components with decay times longer than several hundreds of nanoseconds. The emission spectra peak at around 420 nm for NaI(Tl), CsI(Na) and CaF$_2$(Eu) and at around 540 nm for CsI(Tl). It is often quoted that the number of photoelectrons for CsI(Tl) is smaller than that for NaI(Tl), because the spectroscopic sensitivity of most of the popular bi-alkali photocathodes is better matched to the scintillation spectrum of NaI(Tl) crystals. The KIMS experiment has adapted photomultiplier tubes (PMTs) with green-enhanced photocathodes instead of normal bi-alkali photocathodes to improve the number of photoelectrons. As a result, the number of photoelectrons of the full size crystals was improved to 5–6 per keV, which is similar to that of NaI(Tl) crystals. It is worth mentioning that the use of CsI(Tl) crystals with a silicon photodiode, not only for the space limitation but also for good spectral matching, is preferred in high-energy applications [7, 8].

Among fluorine-based scintillation crystals, CaF$_2$(Eu) is the brightest scintillator. The crystal is formed upon isomorphic substitutions of cations in the host structure by Eu$^{2+}$ ions. The light output is known to be 19 000 photons MeV$^{-1}$ and with a decay time of 0.9 $\mu$s. It has a 424 nm wide emission band that arises from 4f$^6$5d to 4f$^7$ transitions by Eu$^{2+}$ activation.
Table 2. Properties of various scintillation crystals. LY, light yield; $\tau_f$, fast component of decay time; $\lambda_m$, mean wavelength; density; and refractive index are given.

| Crystal      | LY (photons MeV$^{-1}$) | $\tau_f$ (ns) | $\lambda_m$ (nm) | Density | Refractive index | Hygroscopy |
|--------------|-------------------------|---------------|-------------------|---------|-----------------|------------|
| NaI(Tl)      | 38 000                  | 230           | 415               | 3.67    | 1.85            | High       |
| CsI(Tl)      | 65 000                  | 800           | 540               | 4.51    | 1.86            | Slight     |
| CsI(Na)      | 39 000                  | 630           | 420               | 4.51    | 1.84            | High       |
| CaF$_2$(Eu)  | 19 000                  | 940           | 424               | 3.18    | 1.44            | None       |

This emission band matches well with the bi-alkali photocathode sensitivity. The energy resolution was measured to be 9% (full-width at half-maximum (FWHM)) with a 662 keV $\gamma$-ray source [16], and 4 photoelectrons keV$^{-1}$ was achieved at 60 keV energy region with two PMTs [17]. The properties of these crystals, including scintillation characteristics, such as decay times and peak emission wavelengths, are summarized in table 2.

2.2. Quenching and channeling

A nuclear recoil event yields a lower number of scintillation photons than does an electron recoil event for the same energy deposition. Energy calibration of a crystal scintillator is commonly carried out radioactive isotopes emitting $\gamma$-rays of known energies, and therefore the energy measured according to the calibration is often called electron equivalent energy. The ratio of the measured electron equivalent energy to the recoil energy of the nucleus is called the quenching factor. In WIMP searches, one needs to convert the measured energy to the nuclear recoil energy and therefore the quenching factor needs to be measured well.

Measurements of the quenching factors for Na and I recoils in NaI(Tl) crystals have been reported by the Saclay and UKDMC groups [18]–[20]. Quenching factors for Na and I recoils are about 25 and 8%, respectively, and they are independent of the recoil energy, as shown in figure 1. The latest measurement of the quenching factor for Na recoils in NaI(Tl) crystals shows a consistent value of 20–25% [20] with improved errors.

The quenching factor of CsI(Tl) crystals has also been measured by the Saclay [21], UKDMC [22], KIMS [23] and TEXONO groups [24]. The results from all groups are consistent with each other, as shown in figure 2. Cs and I have similar atomic masses and, therefore, quenching factors for them are not measured separately. The quenching factor of CsI(Tl) for nuclear recoils is about 10% at around 100 keV and above, and it increases at lower recoil energies. This behavior is different from NaI(Tl) crystals for which the quenching factor is independent of the recoil energy. The quenching factor for CsI(Na) crystals was measured by KIMS [23] and was found to be smaller than that for CsI(Tl) crystals by a factor of two at lower energies, with rather small energy dependence.

The quenching factors for CaF$_2$(Eu) were measured as 6.9 ± 0.5% and 4.9 ± 0.5% for F and Ca recoils, respectively, by the BPRS collaboration [25]. These were reported to be independent of recoil energy. This result is in contrast with measurements reported by the UK [19, 26] and Osaka [27] groups, which show an increase in the quenching factor at lower energies. Although the trend is similar, the Osaka group’s result showed bigger values, about 10–20% at around 10 keV recoil energy [27].
It is worth noting that details of the experimental conditions, such as the doping fraction, for the measurement of crystal properties, including quenching factors, are not well described in most papers. However, the measured quenching factors from various groups are in fair agreement within the statistical errors. Nevertheless, KIMS showed that the largest systematic uncertainty is from the spread of the measured quenching factors, which attributes about 13% uncertainty to the WIMP–nucleon cross section [50].
A considerable amount of effort has been made to understand the quenching factors. At low energies, the energy loss of recoiling heavy ions in matter can be significantly affected by nuclear collisions (nuclear stopping) in addition to the electronic collisions (electronic stopping). While the main mechanism for producing scintillation light is the electronic energy loss, a large portion of the energy loss is due to nuclear collisions that are generally less efficient in producing scintillation light than are electronic collisions. Lindhard pioneered a theory that calculates the fraction of the energy ultimately transferred to electrons from the initial energy of the ion [28]. Recent studies have tried to explain the measured quenching factors for various scintillating crystals by including the electronic quenching (for example, Birks formula) in the crystals in the Lindhard’s estimation of the so-called nuclear quenching. With this consideration, the estimated quenching factors showed fairly good agreement with experimental data for CsI(Tl) and NaI(Tl) crystals [21, 29].

An interesting point is that there is a factor of two difference between the quenching factors for CsI(Tl) and CsI(Na) crystals at lower recoil energies. This may indicate the importance of electronic quenching, which may depend on the detailed scintillation mechanism of the crystal. Similar considerations provided a better description of recently measured quenching factors for Na recoils in NaI(Tl) crystals, but they are not as good as for the case of CsI(Tl) [20].

In a recent paper [30], a semi-empirical method based on Birks formula and a stopping power estimate to fit measured data with a single parameter was discussed. This method describes quenching factors in a wide energy range for a wide range of scintillators.

Ions entering a crystalline solid along a specific crystallographic axis or plane direction may travel a much longer distance than would be expected based on the normal stopping power. This phenomenon, called channeling, has been known since the 1960s [31]. The DAMA group has suggested that a fraction of the recoil nuclei in their crystals would generate as many photons as recoil electrons without suffering any quenching because of the channeling effect [32]. If this is true, and channeling occurs as frequently as they suggest, the threshold recoil energy would be lower than what is estimated by quenching, at least for some fraction of events. Based on this channeling effect, it is argued that DAMA would be more sensitive to lower mass WIMPs and open some room for consistency with other experiments. More careful study of the effects of channeling on quenching is needed, as this may be very important.

2.3. Pulse shape discrimination (PSD)

WIMP interactions cause nuclear recoils that produce a higher energy loss density than do electron recoils. As a result, the scintillation signal in a nuclear recoil event is significantly quenched, and the pulse shape of a nuclear recoil event tends to be different from that of an electron recoil event. Therefore, discrimination of nuclear recoil signals from the gamma interaction events, which are major internal backgrounds, can be achieved by means of PSD. The most common way of applying PSD is to compare mean times defined by $\langle t \rangle = \frac{\sum A_i t_i}{\sum A_i}$, where $A_i$ is the amplitude of the pulse at time $t_i$. The mean time $\langle t \rangle$ is the same as the decay time constant of the pulse in the cases where the pulse is composed of a single decay component. KIMS improved the PSD effectiveness by fitting each event shape to double exponential decay curves and calculating the mean time of this fitted function. At energies above a few 100 keV, the separation of nuclear from electron recoils can be done very clearly event by event. However, since the resolution of the mean time is highly affected by photon statistics, PSD becomes less effective at low energies. As a result, only a statistical separation is possible in the low-energy region of a WIMP search. The PSD separation power can be described by a quality factor (QF)
Table 3. Background summary of dark matter experiments using crystal scintillators. ‘Bg’ means the background level measured at about 10 keV energy in units of (keV kg day)$^{-1}$.

| Crystal       | Experiment   | Mass (kg) | U (ppt) | Th (ppt) | natK (ppb) | Bg      | Reference |
|---------------|--------------|-----------|---------|----------|------------|---------|-----------|
| NaI(Tl)       | ELEGANT V    | 662       | <500    | <500     | <300       | ~10     | [43]      |
|               | DAMA/NaI     | 100       | 2–10    | 1–6      | ~1.5       | ~1      | [44]      |
|               | DAMA/LIBRA   | 232.8     | 0.7–10  | 0.5–7.5  | ~20        | ~1      | [41]      |
| CsI(Tl)       | KIMS         | 34.8      | 0.75    | 0.38     | 4–5        | ~5      | [50]      |
| CaF$_2$(Eu)   | Tokyo        | 0.31      | ~80     | ~28      | ~5         |         | [17, 36] |

in terms of WIMP search sensitivity [33]. Measurements of QFs have been done by the UKDMC group for NaI(Tl) [19, 20] and by the KIMS and Saclay groups for CsI(Tl) [21, 23]. All these measurements are consistent with each other. The QF of CsI(Tl) is about an order of magnitude higher than that of NaI(Tl). The PSD of CsI(Na) crystals was also studied by KIMS [23], and the QF of CsI(Na) was found to be 2–3 times worse than that of CsI(Tl). The pulse shape of nuclear recoils in CaF$_2$(Eu) is not much different from that of electron recoils and therefore PSD is not applicable to CsF$_2$(Eu) crystals [19].

2.4. Background

Even with extreme care during the preparation of the crystals, internal backgrounds from the remnant radioactive isotopes often become the major limiting factor in low rate counting experiments. The internal and overall background levels for various search experiments are summarized in table 3.

DAMA/LIBRA reported that the residual isotopes in their crystals are 0.5–7.5 and 0.7–10 ppt for $^{232}$Th and $^{238}$U, respectively, with an assumption of secular equilibrium in both Th and U chains. Natural K contamination was reported to be at the level of 20 ppb by using events in which a 1460 keV gamma ray and a 3 keV x-ray are detected in separate crystals in coincidence. They used these events to verify the energy calibration at low energy as well. An interesting observation is that the energy spectrum after the efficiency correction given by DAMA shows a clear peak at around 3 keV. The rate of 3 keV events above the flat background may be explained by the 3 keV energy deposition by the 1460 keV gamma ray escape from the entire array of the crystals. Since this energy region is coincident with that for the annual modulation signature, a better understanding of this background is very important. This could easily be investigated by studying the outer crystals and inner crystals separately, since the escape probability of the 1460 keV $\gamma$-ray is higher for the outer crystals. The background rate of the NAIAD experiment by the UKDMC group was about 7 counts keV$^{-1}$ kg$^{-1}$ day$^{-1}$ at 10 keV.

For CsI(Tl) crystals, the major internal background sources are $^{137}$Cs, $^{134}$Cs and $^{87}$Rb. $^{137}$Cs beta decays primarily to an excited state of $^{137}$Ba. The excited state of $^{137}$Ba emits 662 keV $\gamma$-rays in the transition to its ground state with a half-life of 2.55 min. Therefore, a low-energy beta ray from $^{137}$Cs is detected without the association of additional gamma rays and it can contribute to the low-energy background. Since $^{137}$Cs has a lifetime of 30 years, it does not exist naturally. However, the majority of the CsI(Tl) crystals were contaminated with $^{137}$Cs at the level of 10–100 mBq kg$^{-1}$ [34]. After an extensive study of backgrounds in the crystals,
the KIMS group identified that the $^{137}$Cs contamination originated from the processing water used for the Cs extraction from pollucite, the ore of Cs. By using ultrapure water for powder production, a significant reduction in $^{137}$Cs was achieved. The level of $^{137}$Cs contamination in recently produced crystals was less than 2 mBq kg$^{-1}$ [35].

$^{134}$Cs has a lifetime of 2 years and is mainly produced by the capture of neutrons produced by cosmic ray muons. This contamination dies away once the crystal is stored underground. $^{134}$Cs beta decays to an excited state of $^{134}$Ba followed by an immediate gamma transition to the ground state of $^{134}$Ba. Therefore, background due to $^{134}$Cs can be tagged by additional hits and total energy deposition in the whole array of the crystals.

$^{87}$Rb is a natural isotope with an abundance of 27.84% and it exists in the pollucite. $^{87}$Rb can contribute to the low-energy region since it beta decays with an end point energy of 200 keV, and the reduction in Rb is therefore a critical issue. Using a re-crystallization method, KIMS was able to reduce the Rb contamination to below 1 ppb. It is interesting to note that the $^{40}$K contamination in CsI(Tl) is not measurable.

The major internal background sources in CaF$_2$(Eu) crystals are U and Th contaminations. The ELEGANT-VI group put a lot of effort into the reduction of these, and achieved the background level of 1.2 mBq kg$^{-1}$ for $^{238}$U and 0.11 mBq kg$^{-1}$ for $^{232}$Th [36], with an assumption of secular equilibrium throughout the entire Th and U chains. It found that the major $^{238}$U and $^{232}$Th contaminations were caused by the Eu dopants, even when low background EuF$_3$ powder was used in the crystal production. The CANDLES group will use pure CaF$_2$ crystals, for which it has measured background levels as low as 0.039 mBq kg$^{-1}$ for $^{238}$U and 0.026 mBq kg$^{-1}$ for $^{232}$Th [37]. However, pure CaF$_2$ crystals may have a disadvantage in WIMP searches because their light output is less than one half that of the CaF$_2$(Eu) crystals. Since the ELEGANT-VI group uses pure CaF$_2$ crystal as a light guide for the CaF$_2$(Eu) crystal, it does not need to worry about the $^{40}$K background from the PMT. Using the CaF$_2$(Eu) crystals made of the CANDLES group’s low background CaF$_2$ powder and low radioactive EuF$_3$, the Tokyo group at the Kamioka observatory achieved a background level of 10 counts keV$^{-1}$ day$^{-1}$ kg$^{-1}$ in the 2–10 keV energy region, and a threshold level that is as low as 2 keV, corresponding to 8 photoelectrons [17].

Background from the PMT is unavoidable in crystal scintillator experiments and needs to be well understood. It does not pose a big concern in many experimental conditions. However, in rare event search experiments, the PMT noise requires careful treatment. KIMS tried to take data with PMTs attached to an acrylic box of the same size as the CsI(Tl) crystal. With the same operating and trigger conditions as the WIMP search setup, they took data for about a month. They found a non-negligible number of events with relatively large numbers of photoelectrons within a given time window. The occurrence of these events cannot be explained by the PMT singles rate, which is usually a few kHz. Those PMT backgrounds are largely due to afterpulses in the PMTs. For example, when an accelerated electron interacts with a residual gas molecule or knocks out an ion from the first dynode, it can produce a positive ion that drifts to the cathode and hits the photocathode, generating several electrons. These electrons from the photocathode produce a very fast signal like a single photoelectron but with a large pulse height equivalent to several photoelectrons. The time difference between the initial photon and the afterpulse signal is determined by the velocity of the ions. The time structure of the afterpulse was clearly observed and the residual atoms could be identified [38, 39]. There are other kinds of PMT background that are more difficult to clearly identify. Glasses used for the envelope and the cathode window may be poor scintillators, but may generate signals big enough to be
background at low energies. Even with a very poor scintillation efficiency at the level of $10^{-5}$, several MeV alpha rays from U and Th chain isotopes in the glass can easily generate enough number of photoelectrons to mimic low-energy signals [40]. A quantitative understanding of this background has, however, not been made yet, and the characteristics of this background may be different for different PMTs.

DAMA reported that its PMT noise pulses are mainly rejected by a cut optimized to reject those events with single fast photoelectrons with higher pulse heights [41]. This cut is very effective in rejecting the afterpulse noise induced by ions. DAMA does not mention PMT noise of the second kind, which is more difficult to get rid of. Since the PMTs used for DAMA/LIBRA differ from those used by KIMS, it may not be a matter for direct comparison, but a detailed investigation of such a PMT background may be an important issue.

The UKDMC and Saclay groups have reported background events in NaI(Tl) crystals with pulse decay times that are even shorter than what is expected from the nuclear recoils [18]. These events were recorded even at relatively higher energies, such as several tens of keV, and therefore they cannot originate from PMT noise. By measuring these crystals after un-encapsulating and surface grinding, they concluded that these events originated from alpha decays of radioisotope contaminations on the surface of the crystal [42]. UKDMC also reported similar types of events in CsI(Tl) crystals, and it successfully eliminated them by grinding the surface carefully [42].

### 3. Experiments

There have been several experiments that use NaI(Tl) crystals, including DAMA/NaI, DAMA/LIBRA, UKDMC(NAIAD), ANAIS and ELEGANT V. An experiment using CsI(Tl) crystals has been carried out by the KIMS group. Osaka and Tokyo groups performed experiments using CaF$_2$ crystals. Among these experiments, DAMA/LIBRA has achieved the lowest level of background and accumulated sufficient data to study the annual modulation signature of WIMP interactions. DAMA/LIBRA, without applying PSD, reported the observation of the positive annual modulation signal. On the other hand, other experiments that apply PSD do not observe signature of nuclear recoils from WIMP interactions. It should be noted that other experiments using NaI(Tl) crystals have not achieved background levels as low as DAMA’s. The common internal background and background levels are summarized in table 3 for experiments for which data are available. Results from these experiments are summarized in figures 3 and 4, which show limits on SI and SD WIMP–nucleon interactions, respectively.

#### 3.1. Experiments with NaI(Tl) crystals

Interesting reviews and a brief history of experiments based on NaI(Tl) crystal detectors are given in [55, 56], including early developments by BRS (Beijing-Roma-Saclay). BRS performed experiments in the three underground laboratories, Gran Sasso, Frejus and Mentogou. The first result was reported in 1992 [57] based on 760 g of NaI(Tl) crystals with a background of 5–7 counts keV$^{-1}$ kg$^{-1}$ day$^{-1}$ between 5 and 10 keV.

The UKDMC group reported a fast anomalous component in data that were taken during 1997–2000 with encapsulated NaI(Tl) crystals. Those events with signals faster than even nuclear recoil events were attributed to the surface contamination of alpha decaying isotopes [42]. By de-encapsulating and surface grinding these crystals, it was able to remove
Figure 3. Upper limits on SI WIMP–proton cross section. Results from crystal scintillator-based experiments, NAIAD (NaI(Tl)) [58] and KIMS (CsI(Tl)) [50] are shown by solid lines. The DAMA/LIBRA [49] SI signal region (3σ, with and without channeling considerations) interpreted by [52] is shown as a contour. Also shown are the XENON10 [47] and CDMS [46] results that give strongest bounds. A limit from CoGeNT [51] that constrains the lower mass region is shown as well.

these anomalous components. Using un-encapsulated crystals, the group accumulated a 12523 kg day exposure at Boulby Mine and reported a limit based on pulse shape analysis [58].

An attempt to measure annual modulation with a large array of NaI(Tl) crystals, of total mass 662 kg, was made by the ELEGANT V experiment in the Kamioka underground laboratory. Due to the large background level, it used data at energies above 8 keV, which reduced the WIMP search sensitivity significantly. No sizeable modulation effect was observed with the data obtained for 1 year [43].

DAMA/NaI operated in the Gran Sasso underground laboratory and reported the first limit using the PSD technique with a 4123.2 kg day data sample [59]. A few years later, it reported evidence for an annual modulation from a 4549 kg day exposure [60]. The group accumulated data for 7 years and achieved a total exposure of 57 986 kg day [44]. With this 7-year data, it reports a 6.3σ annual modulation of the signal in the 2–6 keV energy range, but without applying PSD. A fit gives a modulation amplitude equal to 0.0200±0.0032 counts keV⁻¹ kg⁻¹ day⁻¹. Meanwhile, several non-scintillator experiments, such as CDMS [45] and XENON [47], reported results that were incompatible with the interpretation of the DAMA annual modulation as arising from nuclear recoil events by the interactions of commonly quoted WIMPs. Some models, such as inelastic dark matter [48], are not ruled out because the discrepancy might be explained by the differences in experimental techniques and in target nuclei. KIMS [50] also ruled out the DAMA WIMP region with its scintillation crystal containing iodine target.

The DAMA group then developed newer NaI(Tl) crystals and set up a 250 kg mass detector (DAMA/LIBRA). After taking data for 4 years, it announced a new result confirming
its earlier observation of annual modulation [49]. By fitting the combined DAMA/NaI and DAMA/LIBRA data over 11 annual cycles to an annual modulation function, DAMA obtained an amplitude modulation of $0.0223 \pm 0.0027$ $(0.0131 \pm 0.0016)$ for the $2–4$ keV ($2–6$ keV) region with a significance of $8.3$ ($8.2$)$\sigma$. In the measured energy spectrum of DAMA/LIBRA, the event rate above 3.5 keV was reduced by 40% uniformly over DAMA/NaI. This may be understood since the background due to Compton electrons produced by gamma rays from internal sources should have a flat distribution, and the reduction of rate indicates that DAMA/LIBRA crystals have less background. In the region lower than 1 keV, the background looks the same, which again can be understood since the major background in that region is due to PMT noise, and has nothing to do with the improvement in the radiopurity of the crystals. However, between 1 and 3.5 keV, the background shape changed, and one cannot determine easily whether the energy calibration is correct. The background of DAMA/LIBRA at around 2 keV is higher than that of DAMA/NaI, even though the integrated event rates in that region look the same.

As explained in the previous section, if the channeling of recoiled ions occurs as claimed by DAMA, some room at low mass WIMP may be allowed. If the annual modulation observed by DAMA is confirmed by other experiments, and if it is not due to WIMP, it would signal new phenomena that may be more interesting. Therefore, performing a new experiment using NaI(Tl) crystals other than DAMA would be highly desirable. ANAIS is in preparation at Canfranc Underground Laboratory, aiming to build a low background NaI(Tl) crystal detector of 100 kg [62].

Figure 4. Upper limits on SD WIMP–proton cross section. Results from crystal scintillator-based experiments, Tokyo (CaF$_2$(Eu)) [17], NAIAD (NaI(Tl)) [58] and KIMS (CsI(Tl)) [50] are shown by solid lines. The DAMA/LIBRA [49] SD signal region ($3\sigma$, with and without channeling considerations) interpreted by [52] is shown as a contour. Also shown are the COUPP [53] and PICASSO [54] results that constrain the lower mass region.
3.2. Experiments with CsI(Tl) crystals

Studies of the suitability of CsI(Tl) crystal detectors for dark matter searches were made by several groups [21, 24, 61]. CsI(Tl) crystals have some advantages over NaI(Tl) crystals, such as better PSD, a higher quenching factor at low energy, and less hygroscopicity, as discussed above. However, backgrounds from Cs isotopes were found to be very high for the majority of the crystals that were originally available. KIMS developed radiopure CsI(Tl) crystals in a study of material contamination, as described above, and is now the only experiment using CsI(Tl) crystals. It is located in the YangYang underground Laboratory (Y2L) in Korea, where the vertical earth overburden is about 700 m. It reported the first limit using data taken with a single crystal in 2006 [63]. Results from the 3409 kg day data obtained with four crystals with a background level of 5 counts keV$^{-1}$ kg$^{-1}$ day$^{-1}$ were reported in 2007. Results based on PSD analysis to suppress gamma backgrounds further gave strong constraints on the interpretation of the DAMA signal as being due to a WIMP–iodine interaction [50]. Also, the KIMS’ limit on the SD interaction for the case of a pure proton coupling is the most stringent among all direct search experiments at the WIMP mass above 30 GeV, while COUPP, and more recently PICASSO, both based on superheated liquids containing $^{19}$F, achieved the most stringent limit for the WIMP mass below 30 GeV [53, 54]. Limits on the SD WIMP–proton cross section are summarized in figure 3. It is worth noting that the current front-running experiments, such as CDMS and XENON, are not sensitive to the SD interaction for the case of a pure proton coupling since their target nuclei contain even numbers of protons. Therefore, KIMS is complementary to these experiments. It has been suggested that an independent constraint on SD coupling to proton is necessary to constrain models [64]. KIMS is currently taking data with 12 crystals, with a total mass of 104 kg. After stable data taking for more than a year, KIMS should be able to test for annual modulation.

3.3. Experiments with CaF$_2$(Eu)

CaF$_2$(Eu) crystals have been also investigated as dark matter detectors because $^{19}$F is a highly sensitive target for SD interactions of WIMPs. The first dark matter search result with CaF$_2$(Eu) was published by the BPRS collaboration [25]. This used 370 g of crystal with a background level higher than 10 counts kg$^{-1}$ keV$^{-1}$ day$^{-1}$. The Osaka group has performed a search experiment at the Oto underground laboratory using 2.6 kg of CaF$_2$, but with a somewhat higher background level [65]. The Tokyo group has also carried out an experiment at the Kamioka Observatory with 310 g of CaF$_2$(Eu) crystals. It identified the major background below 10 keV as coming from Cherenkov photons in the quartz light guides between the crystals and PMTs, which is relatively easy to reject. After removing these background events by a pulse shape analysis, the group was able to reduce the background to a level less than 10 counts keV$^{-1}$ kg$^{-1}$ day$^{-1}$. Using this data set, it set a limit on SD interactions, which is also shown in figure 3 [17].

4. Prospects

Although there has been significant advancement in experimental techniques in cryogenic detectors, noble gas liquid scintillators and other alternatives, the crystal scintillators still offer an attractive method for dark matter searches. One of the virtues of crystal scintillators is their
simplicity of operation, which does not require a complex cryogenic system that is expensive to maintain stably over a long-term operation of large-scale detectors. The relatively lower cost compared with the cryogenic technique, both for construction and for operation of a large mass detector, is also an attractive point. The annual modulation reported by DAMA needs to be confirmed or denied by other experiments, regardless of its origin. If the modulation is confirmed by other experiments and if it is not due to WIMP interactions, it would still be very interesting. Unfortunately, DAMA/LIBRA cannot help much to resolve this conflict as its modulation effect is already more than 8 standard deviations, and further data taking will not improve this very much. Therefore, independent experiments that confirm or reject the claimed annual modulation are very important. At the moment, KIMS with 100 kg of CsI(Tl) crystals is in a good position to explore this possibility. It would also be worthwhile to have another group, such as ANAIS, carry out an experiment with more than 100 kg NaI(Tl) crystals with the same level of radiopurity as the DAMA crystals.

In order to go beyond the current scale of detector mass for crystal scintillator dark matter searches, one needs breakthroughs in a couple of areas. Further reduction of the internal background—for example, by purifying the powder using even higher purity water—is one area. An improvement in photon counters is another avenue to be pursued. The recent development of higher-quantum-efficiency photocathodes by Hamamatsu is very promising in this regard. It has already produced a series of PMTs with a quantum efficiency as high as 40% using a so-called ultra bialkali (UBA) photocathode [66]. A similar effort to enhance the quantum efficiency at longer wavelengths that match the emission spectrum of CsI(Tl) is in progress. Higher quantum efficiency will improve the sensitivity of WIMP searches by reducing the threshold recoil energy as well as improving the PSD capability, which is mainly limited by the photon statistics. Another PMT improvement could be the use of metal-packaged envelopes that reduce K contamination near the crystals.

5. Conclusion

We have reviewed the crystal scintillation detectors for direct dark matter search experiments. Since the crystal scintillators have the advantages of easy operation and lower cost than the cryogenic technique for large mass detectors in terms of both fabrication and operation, they have been attractive tools for direct dark matter searches. An urgent immediate use for crystal scintillators in dark matter searches is to directly test the DAMA signal for an annual modulation. The KIMS experiment based on CsI(Tl) crystals continues to take data, and the ANAIS experiment using NaI(Tl) crystals is being pursued. With further reduction in radioisotope contamination and improvements in photon counting devices, crystal scintillation detectors could finally clarify the origin of the DAMA modulation signal.

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